Renewable biomass-derived carbons for electrochemical capacitor applications

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Funding information
National Natural Science Foundation of China, Grant/Award Numbers: 52062012, 51525206, 520105010, 21603048; Key Science & Technology Project of Hainan Province, Grant/Award Number: ZDYF2020028

Abstract
Biomass is rich, renewable, sustainable, and green resources, thereby excellent raw material for the fabrication of carbon materials. The diversity in structure and morphology of biomass are relevant in obtaining carbon materials with different structures and performances. The inherent ordered porous structure of biomass also benefits the activation process to yield porous carbons with ultra-high specific surface area and pore volume. Besides, obtained biomass-derived carbons (BDCs) are hard carbon with porous morphology, stable structure, superior hardness/strength, and good cycling performances when used in electrochemical capacitors (ECs). The inherent N, S, P, and O elements in biomass yield naturally self-doped N, S, P, and O BDCs with unique intrinsic structures. In this paper, the synthesis approaches and applications of BDCs in ECs are reviewed. It shows that BDCs electrochemical performances are highly determined by their pore structures, specific surface areas, heteroatoms doping, graphitization degree, defects, and morphologies. The electrochemical performances of BDCs can further be improved by compositing with other materials, such as graphene, carbon nanofibers/nanotubes, transition metal oxides or hydroxides, and conducting polymers. The future challenges and outlooks of BDCs are also provided.

Keywords
biomass-derived carbons (BDCs), electrochemical capacitors (ECs), pore structure, specific surface area

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The increasing human population and the development of social economy require enhanced energy consumption. Fossil fuel-based energy has several adverse effects on the environment, including global climate change, loss of biodiversity, and pollution of air, water, and marine. Therefore, tremendous efforts have been devoted to reduce fossil fuel consumption and explore low-cost, abundant, environmentally friendly, sustainable, and renewable energy resources.  

Energy may be classified into two main categories. The first consists of nonrenewable energy, such as coal, petroleum, and natural gas. The second deals with renewable energy, such as geothermal, solar, wind, tide, and biomass. However, renewable energy based on solar, wind, geothermal, and tide energies is often difficult to apply to large scale for long-time use due to its regional, intermittent, and unstable features. Consequently, the efficient energy conversion and storage technologies are required. The most useful electrochemical energy storage devices are based on rechargeable batteries and electrochemical capacitors (ECs). Rechargeable batteries, such as lithium-ion batteries, store energy via faradaic reactions. These devices have been supplying electric power for our daily appliances, such as cellphones, laptops, and electric vehicles for quite some time now. Rechargeable batteries usually possess high energy density but still suffer from low power density. By comparison, ECs possess high power density and long cycle life but still limited by low energy density when compared to batteries. 

Based on the charge storage mechanisms and device characteristics, ECs can be classified into three categories: (i) electric double-layer capacitors (EDLCs), (ii) pseudocapacitors, and (iii) asymmetric capacitors. Pseudocapacitors store electric charge through surface-controlled faradaic reactions because pseudocapacitive materials usually trigger fast and highly reversible redox/intercalation reactions on the electrode surface or near-surface. Asymmetric capacitors can be further classified into two types, that is, devices consist of two capacitive electrodes or hybrid capacitors. Hybrid capacitors like battery-type capacitors are identified as a kind of device in which one electrode stores electric charge through battery-type faradaic process, while the other electrode stores electric charge based on capacitive mechanism. Generally, hybrid capacitors combine the advantages of high-power density of EDLCs and high-energy density of pseudocapacitors, yielding outstanding comprehensive performances. However, the cycling stability of such devices still requires further strengthening. By comparison, EDLCs store energy via electrostatic interaction by reversibly adsorbing/desorbing cations and anions from the electrolyte at the electrode/electrolyte interface to form the electric double layer (EDL). From the point of view of electrode materials, pseudocapacitors or partial asymmetric capacitors can be pseudocapacitive materials that deliver higher specific capacitance, while for EDLCs are mainly carbon-based materials, which exhibit better cycling stability and rate performance. Carbon-based materials like porous carbons have aroused much attention thanks to their various advantages. Porous carbons, such as activated carbon, carbon nanotubes, porous carbon nanofibers, and graphene have been widely explored due to their elevated specific surface area, adjustable porous structure, excellent electrical conductivity, and superior physicochemical stability.

Carbon-based electrode materials not only are excellent candidates for ECs and hybrid capacitors, but also play an important role in supporting active materials of pseudocapacitors. Alternatively, biomass-derived carbons (BDCs) are excellent materials for ECs due to many features, such as sustainability, renewability, abundant natural sources, and low cost. This paper is to review the recent advances in the preparation of BDCs from different biomass precursors. Note that the electrochemical properties of ECs are highly involved in the characteristics of BDCs, including morphology, specific surface area, pore structure, heteroatoms doping, graphitization degree, and defects (Figure 1). Moreover, the electrochemical performances of BDCs can further be improved by compositing with other materials, such as graphene, carbon nanofibers/nanotubes, transition metal oxides or hydroxides, and conducting polymers.
2 | CHARGE STORAGE OF EDLCS

The charge storage of EDLCs has first been described by Hermann von Helmholtz. This pathway occurs through electrostatic adsorption of ions at the interface between the electrode and electrolyte under certain voltage simultaneously proceeding on both electrodes. In Gouy–Chapman model, nonuniform distribution of ions (anions and cations) takes place due to diffusion and dipole movement of solvent molecules driven by thermal fluctuation. To consider highly concentrated electrolyte solutions, Stern has introduced two regions of ion distributions, namely, the compact layer and diffusion layer. However, the Stern model suffers from limitations at elevated applied voltages, highly concentrated electrolytes, or in ionic liquids because it ignores the effects of ions interaction. Later, Kornyshev et al developed the Landau–Ginzburg-type continuum theory by considering the EDL structure in ionic liquids different from that in solvent-containing electrolytes. In ionic liquids, the absence of solvent molecules for screening cations and anions would lead to strong ions interactions. Therefore, EDL would create an overscreening effect under lower potentials (0.26 V) but display the crowding effect under larger polarization potentials (2.6 V) attributed to the different degrees of ionic interactions.

The classical EDL theory states proportionality between the specific capacitance of EDLCs and specific surface area of the electrode. However, this might be different for porous carbon because the pore structure is closely related to the specific capacitance as ions can be desolvated and stored in micropores. In other words, the classical two-dimensional (2D) planar EDL model cannot explain the mechanism of charge storage of three-dimensional (3D) porous carbon because 2D model does not consider surface curvature and porous effects. The previous research reported that the capacitance would rapidly increase for pore size less than 1 nm, especially the optimal capacitance will obtain when pore size decreases to 0.7 nm in aqueous electrolytes and 0.8 nm in organic electrolytes. Hence, the pore size and pore size distribution can impact the capacitance besides the specific surface area. For instance, Cl– and ClO4– can be absorbed in carbon molecular sieve, while SO42– cannot.

3 | BDCS APPLICATION IN ECS

Porous carbon materials are excellent candidates for ECs due to their high specific surface area, abundant porous structure, superior physicochemical stability, and outstanding electrical conductivity. But the conventional carbon materials have the inherent disadvantages. For instance, carbon nanotubes exhibit superior rate capability thanks to their outstanding electrical conductivity, but the insufficient utilization of inner surface area may lead to moderate specific capacitance. Graphene is made of 2D carbon monolayers, making its promising electrode material for EDLCs. However, restacking of individual sheets always generates irreversible agglomeration, thereby reducing the specific surface area. Although substantial progress has so far been achieved for carbon nanotubes and graphene, meeting the requirements of large-scale energy storage applications is still challenging. Therefore, the preparation of low-cost, renewable, sustainable, and environmental friendliness electrode materials for ECs is highly desirable.

Biomass is the most popular precursor of porous carbon materials due to its large abundance, sustainability, renewability, low cost, and nontoxicity to the environment and human health. Furthermore, biomass possesses unique micro-/nanostructure and ordered intrinsic pores, beneficial for the preparation of activated porous carbon with high specific surface area and large pore volume. In particular, naturally ordered porous structure of biomass consisting of inherent rich pore structures and aligned microchannels is found in most biomass, such as coconut shells and natural woods, and might provide adequate diffusion channels for the activator to readily prepare porous carbon with high specific surface area. Moreover, BDCs are much different from petroleum coke and coal asphalt-based carbon thanks to their randomly oriented nanotextures and graphite microcrystalline disorderly stacking, thereby providing large numbers of voids and defects. These features should benefit subsequent activation processes and create large amounts of pore...
structures. Also, the hard carbon structure of BDCs is relatively stable when compared to soft carbon under high power and voltage conditions.

In most cases, BDCs may inherit or evolve the special structures of biomass precursors. The diversity of biomass leads to the formation of BDCs with spherical, fibrous, sheet-like, and skeleton structure. Biomass, especially lignocellulosic biomass, consists of mainly cellulose, hemicellulose, and lignin (Figure 3), chemically composed of C, H, and O with the partial presence of N, S, and P, as well as a trace of Fe, Ca, K, Si, and other elements. The constituents of lignocellulose are strongly intermeshed and held by covalent or noncovalent forces. Cellulose is a homopolymer consisting of β-D-glucopyranose units linked by β-glycosidic bonds and makes up 40%–50% lignocellulosic biomass. Hemicellulose accounts for 25%–35% biomass with branching and amorphous features, as well as a low degree of polymerization. Thus, hemicellulose can easily be decomposed under thermal conditions. Lignin is also a major component of lignocellulosic biomass and amorphous polymer consisting of 15%–20% biomass. Besides, lignin is rich in aromatic functionality consisting of p-coumaryl alcohol, coniferyl alcohol, and sinapyl alcohol. Biomass and its byproducts have widely been utilized as renewable and sustainable precursors of BDCs, including coconut shells, wheat straw, lotus leaves, waste bones, and yeast.

### 4 | PREPARATION OF BDCS

Most biomass raw materials cannot directly be used as electrode materials in ECs due to their undesirable impurities, low specific surface areas, and underdeveloped porous
structures. Thus, various synthesis methods, such as pyrolysis, hydrothermal, and activation, have been utilized to increase the specific surface area and porosity, as well as regulate the surface chemistry and morphology of BDCs. The reason for this has to do with electrochemical performances and morphologies of BDCs, which highly rely on the intrinsic structures of biomass precursors and the synthesis methods. Figure 4 shows the scheme of the preparation processes of BDCs from various biomass raw materials. Some examples of BDCs synthesized by different methods using various biomass raw materials and activators are also summarized in Table 1.

Carbonization mainly includes pyrolysis and hydrothermal carbonization. Biomass gradually converts into biocarbon during pyrolysis by thermochemical decomposition at high temperatures in the absence of oxygen. Note that high-temperature pyrolysis includes conventional pyrolysis, microwave pyrolysis, solar pyrolysis, plasma pyrolysis, and catalytic pyrolysis.

Besides high-temperature pyrolysis carbonization, hydrothermal carbonization is also a thermochemical process allowing the conversion of biomass precursors into hydrocarbons at relatively low temperatures (180~250 °C) and saturated pressure (2~10 MPa) within enclosed space. The hydrothermal process generally involves dehydration, polymerization, and carbonization. During this process, the chemical structure of hydrocarbons, such as crosslinks of aromatic polymer, organic functional groups, surface porosity, and ultimate components, can be tailored by the hydrothermal carbonization parameters, such as temperature, time, residence, substance concentration, and presence of catalysts.

BDCs with excellent properties often require more than pyrolysis or hydrothermal carbonization. Examples include activation processes allowing the creation of pronounced pore structures, such as physical activation, chemical activation, physicochemical activation, microwave-assisted activation, and template-assisted activation.

Physical activation mainly utilizes water steam and CO\(_2\) as activating agents to prepare BDCs. The common preparation process involves carbonization at 300~800 °C followed by activation at 700~1000 °C using the chemical reaction between carbon and H\(_2\)O (Equation (1)) or CO\(_2\) (Equation (2)) to create porous BDCs:

\[
C + H_2O \rightarrow H_2 + CO \quad \Delta H = +132 \text{ kJ} \cdot \text{mol}^{-1},
\]

\[
C + CO_2 \rightarrow 2CO \quad \Delta H = +173 \text{ kJ} \cdot \text{mol}^{-1}.
\]
Table 1: BDCs synthesis by different preparation methods

| Carbonization        | Activation          | Activators | Biomass raw materials                                                                 |
|----------------------|---------------------|------------|---------------------------------------------------------------------------------------|
| Pyrolysis carbonization | Chemical activation | KOH        | Alfalfa flowers,41 Wheat straw,61 Bamboo,65 Pine sawdust,66 Loofah sponge,67 Rice husk,68 Garlic skin,50 Pea protein,69 Rose,62 Lotus root shell,70 Tea waste,71 Leftover rice,72 Borassus flabellifer flower,73 Peanut shell,78 Houttuynia,76 Pinecone,76 Wood tar,77 Walnut shells,79 Switchgrass,79 Coconut shells50 |
|                      |                     |            | K₂FeO₄,50 Bamboo char,56 Leaves of phoenix tree,82 Loofah83 NaOH,55 Mangosteen peel,54 Peanut shell/Corn cob/Wheat bran/Rice husk,69 Peanut shell90 ZnCl₂,59 Polar catkins,63 Waste tea,68 Glucose,69 Juncus,66 Nori90 |
|                      |                     |            | C₆H₁₅NO₃,60 ZnO,57 Plane tree bark91 H₂PO₄,64 CH₄COOK,75 Switchgrass79 KOH/HNO₃,78 Beancurd52 KOH/KNO₃,77 Cotton91 HNO₃/NaOH,69 Withered rose flower94 KOH/Li₂CO₃,61 Lotus leaves62 ZnCl₂/KCl,74 Cotinus coggygria flower95 |
|                      |                     |            | Physical activation                                                                 |
|                      |                     |            | Steam,97 Sugarcane bagasse/Coconut shell/Endocarp of babassu coconut,96 pine nutshell,99 Barley straw100 Onion peels103 |
|                      |                     |            | CO₂,103 Hybrid willow,101 Barley straw,100 Sargassum fusiforme102 |
|                      |                     |            | Air,102 Coconut shells,97 Sugarcane bagasse/Coconut shell/Endocarp of babassu coconut,96 pine nutshell,99 Barley straw100 Onion peels103 |
| Physical activation  |                     |            | Physicochemical activation                                                             |
|                      |                     |            | FeCl₃+ZnCl₂/CO₂,97 Peanut shell,104 FeCl₃+MgCl₂/CO₂,97 Peanut shell,104 ZnCl₂+MgCl₂/CO₂,97 Peanut shell,104 Steam/KOH,97 Lignin105 FeCl₃/CO₂,97 Lignite106 |
|                      |                     |            | Microwave-assisted activation                                                          |
|                      |                     |            | Air,107 Human hair,107 Crude lignin,108 Camellia oleifera shell,109 Waste palm,110 Camellia oleifera111 |
|                      |                     | KOH        | KOH,107 Crude lignin,108 Camellia oleifera shell,109 Waste palm,110 Camellia oleifera111 |
|                      |                     |            | CO₂,107 Coconut shell112 Steam,107 Coconut shell112 Steam/CO₂,107 Coconut shell112 |
|                      |                     |            | Templated-assisted activation                                                          |
|                      |                     | NaCl/KCl   | Kitchen waste hydrolysis residue113 Glutinous rice114 Cotton115 |
|                      |                     | Mg(OH)₂    | MgO/ZnCl₂,113 Glutinous rice114 Cotton115 |
|                      |                     |            | Hydrothermal carbonization                                                             |
|                      |                     | KOH        | Cypress coats,116 Pomelo peel,117 Wood fibers118 Garlic peel,119 Bamboo shoot shells,120 Crab shells/rice husks,121 Coconut shell/Sewage sludge,122 Composting leachate121 |
|                      |                     |            | Microwave-assisted activation                                                          |
|                      |                     | KOH        | Distiller’s dried grains with solubles124 |

The activation process involves controlled active carbon atom burn-off and elimination of volatile matters. The quality of BDCs greatly depends on the reaction parameters, such as activation temperature, activation time, gas flow rate, and presence of catalysts, among others. In addition to carbonization and activation parameters, different biomass materials also require appropriate activators to obtain BDCs with high specific surface areas and well-developed pores. The reason for this has to do with the variable structures and compositions of different biomass materials. For instance, BDC from willow with specific surface area of 738.74 m²·g⁻¹ and pore volume of 0.37 cm³·g⁻¹ can be obtained through CO₂ activation at 800 °C.101 Similarly, BDC from cattail with specific surface area 441.12 m²·g⁻¹ and pore volume of 0.33 cm³·g⁻¹ can be prepared by CO₂ activation at 800 °C.140 Apart from CO₂ activation, water steam is also widely employed as an effective activator, especially in industry due to its low cost, cleanness, and high safety. Examples include BDC from pine nutshell obtained through water steam activation at
850 °C with specific surface area reaching 956 m²·g⁻¹ and mesopore ratio of 37.1%.

Nevertheless, BDCs with high specific surface area above 2000 m²·g⁻¹ by physical activation are still difficult. Alternatively, chemical activation is often used to prepare BDCs with well-developed micropores and ultrahigh specific surface areas. Various types of activators are utilized to this, including KOH, NaOH, ZnCl₂, HNO₃, and H₃PO₄. Among these, KOH has shown the best activation effect for increasing the specific surface area, thereby being the most widely used activator. Almost all carbon precursors based on hard or soft carbon can be activated by the extreme KOH etching effect during the reaction between KOH and carbon at higher activation temperature (> 750 °C, Equations (3)–(5)). During this process, K⁺ diffuses into bulk of the carbon skeleton to expand or create new pores during the activation process.141,142

\[
\begin{align*}
6\text{KOH} + 2\text{C} & \leftrightarrow 2\text{K} + 3\text{H}_2 + 2\text{K}_2\text{CO}_3, \\
\text{K}_2\text{CO}_3 & \leftrightarrow \text{CO}_2 + \text{K}_2\text{O}, \\
\text{K}_2\text{CO}_3 + 2\text{C} & \leftrightarrow 2\text{K} + 3\text{CO}.
\end{align*}
\]

Various types of biomass exhibit different specific surface area and pore volume after chemical activation. For instance, BDC from pine sawdust by KOH activation possess yields high specific surface area up to 2330.89 m²·g⁻¹ and large pore volume of 1.914 cm³·g⁻¹.66 BDC from tobacco rods by hydrothermal carbonization and KOH activation holds large specific surface area of 2115 m²·g⁻¹ and pore volume of 1.22 cm³·g⁻¹.143 Others BDCs, such as bamboo (2221.1 m²·g⁻¹)65 and rice husk (2804 m²·g⁻¹)148, are characterized by elevated specific surface areas after KOH activation. Compared to KOH, NaOH activation possesses are advantageous in terms of less corrosion, lower weight dosage, and low cost.144 NaOH is also widely employed for the preparation of porous carbon. For example, BDC from wheat bran by NaOH activation at 800 °C has shown substantial specific surface area (2543 ± 89 m²·g⁻¹) and large total pore volume (1.684 cm³·g⁻¹).145 BDC obtained from coconut shell also holds large specific surface area (2825 m²·g⁻¹) and superior total pore volume (1.498 cm³·g⁻¹) after NaOH activation.144 Compared to KOH and NaOH, ZnCl₂ is a milder activator, meaning that lower activation temperature is required to prepare BDCs.90,146 The activation mechanism is possibly based on the catalytic dihydroxylation and dehydroxylation during pyrolysis, in which H and O are released from biomass in the form of H₂O to form rich microporous structures. Meanwhile, ZnCl₂ could provide a framework for carbon deposition during the activation process. In the subsequent step of acid washing, ZnCl₂ would leave the voids of the carbon skeleton to yield additional porosity.115

In this respect, BDC from pine cone and treated by ZnCl₂ activation at 500 °C for 2 h displays high specific surface area of 2771 m²·g⁻¹ and large mesopore volume of 1.22 cm³·g⁻¹.147 Similarly, BDC obtained from oleaster fruit has shown specific surface area of 1719 m²·g⁻¹ and total pore volume of 1.193 cm³·g⁻¹ after ZnCl₂ activation at 450 °C for 1 h.146

Other activation methods like physicochemical activation, templated assisted activation, and microwave assisted activation have also been applied in the preparation of BDCs (Table 1). Note that despite the advantages of chemical activation for preparing BDCs with high specific surface area and well-developed pores, the method still suffers from the environmental pollution and equipment corrosion, limiting its wide applications. Thus, the development of economical, efficient, simple, and safe activation routes is highly desirable to solve these issues. Self-activation can convert biomass to BDC without the additional activators because biomass with inherent inorganic salts or other metal ions (such as K, Na, Fe, and Ca) could etch carbonized precursors or react with gases (such as CO₂, H₂O, and H₂) emitted during carbonization.148,149 This self-activation route has so far been applied for preparing BDCs from coconut shell, almond stone, pecan shell, and slash pine sawdust.150 Similarly, most animal bones, such as cattle bones, are composed of collagen and hydroxyapatite (HA, Caₓ(PO₄,CO₃)y(OH)). These materials can easily convert into BDCs through direct carbonization without any additional templates or activators thanks to the in-situ HA-induced self-activation.151

5 | INFLUENCING FACTORS OF BDCS IN ECS

The electrochemical performances of ECs highly depend on the characteristics of BDCs electrode materials, including pore structure, specific surface area, heteroatoms doping, graphitization degree, defects, and morphologies. The electrochemical performances of BDCs can further be improved by compositing with other materials, such as graphene, carbon nanofibers/nanotubes, transition metal oxides or hydroxides, and conducting polymers. Figure 5 depicts the influencing factors of BDCs and obtained composites in ECs, and Table 2 summarizes the BDCs used in ECs based on different recent biomass precursors.

5.1 | Pore structure and specific surface area

As mentioned above in Section 2, micropores especially pore size smaller 1 nm are the most effective pore size for
ions storage, where effective micropore distribution would generate preferable capacitance because micropores provide large surface area for ions absorption/desorption. However, excessively increased micropore volume would rise the transfer resistance of ions into porous carbon inner channels, thereby declining the rate performance during charge/discharge cycling at high current densities. Mesopores often provide fast ions transport pathways from the bulk of the electrolyte to the inner active sites of electrode materials, leading to excellent rate performance and power density. Macropores could store affluent ions as ion-buffering reservoirs, thereby reducing the ions transport distance and further improving the electrochemical performances.

BDCs with dominant micropores often possess substantial specific surface area and smaller pore size, useful for elevated specific capacitance and energy density. For example, micropore dominant BDC from flaxseed residue shows specific surface areas reaching up to 3230 m²·g⁻¹ and micropore specific surface area of 3046 m²·g⁻¹. Besides, more than 70.1% of micropore volume is shown to originate from the large micropores (1–2 nm), useful to induce high specific capacitance values of 369 and 398 F·g⁻¹ in KOH and H₂SO₄ electrolytes, respectively, as well as a

| Biomass                  | \( S_{\text{BET}} \) (m²/g) | \( S_{\text{micro}} \) (m²/g) | \( V_{\text{total}} \) (cm³/g) | \( V_{\text{micro}} \) (cm³/g) | Doped heteroatoms | Specific capacity (F/g) (cell type) | Electrolyte | Current density (A/g) |
|--------------------------|-------------------------------|--------------------------------|-------------------------------|-------------------------------|-------------------|-----------------------------------|--------------|-----------------------|
| Pine sawdust             | 2331                          | 543                            | 1.914                         | 0.975                         | O, N, S           | 175.6 (3E)                       | 1M KOH       | 0.5                   |
| Coconut shells           | 3513                          | 2                             | 1.1                           | 0.8                           | O                 | 325 (2E)                         | 6M KOH       | 0.1                   |
| Bamboo stalk             | 1732                          | 0.97                          | 0.8                           | 0.8                           | O                 | 222 (3E)                         | 6M KOH       | 0.5                   |
| Lotus leaves             | 2351                          | 1.37                          | 0.52                          | 0.52                          | N, O              | 478 (3E)                         | 6M KOH       | 1                     |
| Cypress coats            | 1326                          | 0.779                         | 0.347                         | N, O                          | 345 (3E)          | 330 (3E)                         | 1M H₂SO₄     | 1                     |
| Rice husk               | 2804                          | 1.797                         | 0.476                         | 0.476                         | O                 | 278 (2E)                         | 6M KOH       | 0.5                   |
| Garlic skin              | 2818                          | 1.327                         | 0.37                          | 0.37                          | O                 | 427 (3E)                         | 6M KOH       | 0.5                   |
| Clover stems             | 2244                          | 1.44                          | 0.5                           | 0.5                           | O, N              | 436 (3E)                         | 1M H₂SO₄     | 1                     |
| Pea protein              | 3500                          | 1.64                          | 1.16                          | 1.16                          | N, O              | 413 (3E)                         | 1M KOH       | 1                     |
| Wood fibers              | 1807                          | 0.836                         | 0.775                         | 0.775                         | N, O              | 345 (3E)                         | 6M KOH       | 0.5                   |
| Lotus root shell         | 2961                          | 1.47                          | 0.21                          | 0.21                          | O                 | 345 (3E)                         | 2 M KOH      | 0.5                   |
| Chitosan                 | 2204                          | 1.19                          | 1.04                          | 1.04                          | O, N, P           | 317 (3E)                         | 6M KOH       | 0.5                   |
| Shaddock endotheliums    | 1265                          | 0.746                         | 0.583                         | 0.583                         | N, O, S           | 550 (3E)                         | 1M H₂SO₄     | 0.2                   |
| Leftover rice            | 2184                          | 1.839                         | 0.356                         | 0.356                         | O                 | 153.2 (2E)                       | 1M TEABF₄/PC | 0.2                   |
| Mangosteen peel          | 2623                          | 7.31                          | 0.31                          | 0.31                          | O                 | 357 (3E)                         | 6M KOH       | 1                     |
| Beancurd                 | 1735                          | 1.10                          | 0.82                          | 0.82                          | N, O              | 315 (2E)                         | 1M H₂SO₄     | 0.1                   |
| Cotton                   | 1508                          | 0.1                           | 0.1                           | 0.1                           | O, P              | 278 (3E)                         | 6M KOH       | 1                     |
| Phoenix tree leaves      | 2208                          | 1.27                          | 1.0                           | 1.0                           | O, N              | 273 (3E)                         | 1M H₂SO₄     | 0.5                   |
| Lignin                   | 3065                          | 2.05                          | 0.73                          | 0.73                          | O                 | 325 (3E)                         | 6M KOH       | 0.5                   |
| Houttuynia               | 2090                          | 1.1                           | 0.92                          | 0.92                          | N, O              | 473.5 (3E)                       | 6M KOH       | 1                     |
| Wood tar                 | 2490                          | 1.28                          | 0.32                          | 0.32                          | O                 | 338.5 (3E)                       | 6M KOH       | 1                     |
| Juncus                   | 1380                          | 1.163                         | 1.163                         | 1.163                         | O, N              | 290.5 (3E)                       | 6M KOH       | 0.5                   |
| Palm loofah fibers       | 2000                          | 1.35                          | 1.35                          | 1.35                          | O                 | 337 (2E)                         | 1M NaCl      | 1                     |
| Walnut shells            | 3577                          | 2.19                          | 2.19                          | 2.19                          | O                 | 330 (2E)                         | 6M KOH       | 0.1                   |
| Plane tree bark          | 1512                          | 2.28                          | 0.32                          | 0.32                          | O                 | 286 (3E)                         | 6M KOH       | 0.5                   |
| Eleocharis dulcis        | 2454                          | 1.345                         | 0.65                          | 0.65                          | O, P, N           | 340.2 (3E)                       | 6M KOH       | 1                     |
| Flaxseed residue         | 3230                          | 1.67                          | 1.41                          | 1.41                          | O                 | 369 (3E)                         | 6M KOH       | 0.5                   |
| Elm seeds                | 2598                          | 0.5                           | 0.5                           | 0.5                           | O                 | 501 (3E)                         | 6M KOH       | 1                     |
FIGURE 5 Influencing factors of BDCs and its composites in ECs

substantial energy density of 61.2 Wh·kg⁻¹. BDC from mango skins provides a specific surface area of 2776 m²·g⁻¹ and micropore specific surface area of 1600 m²·g⁻¹. The large micropore specific surface induces a maximum specific capacitance up to 493 F·g⁻¹ and energy density of 27.5 Wh·kg⁻¹. Moreover, BDCs with dominant mesopores often exhibit excellent ions transmission performances because mesopores can serve as ions diffusion channels. For instance, BDC from coconut shells with 75% Vₘeso/V₅ሠ ratio without any conductive agents yields low equivalent series resistance and excellent rate performance as the large mesoporosity that benefits ions transmission and conductivity.

On the other hand, elevated specific surface area BDCs may not necessarily lead to higher specific capacity. A typical example for that lies in BDC from fungus (Auricularia), which shows porous graphene-like carbon after hydrothermal treatment in KOH solution followed by carbonization. However, its specific surface area of 1103 m²·g⁻¹ is lower than that of the material prepared by hydrocarbon followed by KOH activation (1543 m²·g⁻¹). The former material can deliver higher gravimetric (volumetric) capacitance of 374 F·g⁻¹ or 360 F·cm⁻³ than latter (280 F·g⁻¹ or 230 F·cm⁻³). The interconnected hierarchical porous structure and substantial specific surface area of BDC from fungus (Auricularia) lead to excellent specific capacitance, rate performance, and cycling stability. Hence, the hierarchical porous structure together with the high specific surface area is useful for improving the electrochemical performances.

Similarly, BDC from wheat straw shows different specific surface area and pore structure characteristics after treatment with different contents of KOH activation. The substantial specific surface area mainly originates from the micropores and smaller volume ratio of mesopores to micropores, not conducive to the penetration of ions. Consequently, BDC from wheat straw displays the highest specific surface area of 2560 m²·g⁻¹, but lower specific capacitance. This suggests that elevated specific surface area combined with better pore size distribution should enhance the electrochemical performances. Hence, the construction of appropriate hierarchical porous structures containing micropores, mesopores, and macropores would be an effective strategy to improve the electrochemical performance and energy storage in ECs.

BDCs with hierarchical porous and micro-/meso-/macropores provide large specific surface area to accommodate ions, as well as abundant multiple channels for ions transmission and affluent reservoirs space for ion-buffering. Therefore, BDCs with ordered pore structures, narrow pore size distributions, and interconnected hierarchical porous frameworks can improve the electrochemical performances. In Figure 6A, BDCs with hierarchical pores obtained from mixed of rice husks and crab shells treated by the hydrothermal carbonization and KOH activation show high specific surface area reaching up to 3557 m²·g⁻¹ and well-developed hierarchical porosity containing micropores, mesopores, and macropores (Figure 6B–F). The reason for this has to do with the inorganic and synergy between rice husks (SiO₂) and crab shells (CaCO₃) combined with KOH etching effect. As a result, the obtained BDCs are characterized by excellent capacitance retention, good cycling stability, and elevated energy density and power density. Also, BDC with hierarchical egg-box like from moringa oleifera branches reveals superior specific surface area (2312 m²·g⁻¹), large pore volume (1.2 cm³·g⁻¹), unique egg-box-like frameworks, and interconnected hierarchical porous structure. In turn, these features translate into high specific capacitance, excellent rate performance, and good cycling stability.

5.2 Heteroatoms doping

Heteroatoms doping by elements, such as O, N, S, P, and F, can enhance the electrical conductivity, change the surface wettability, introduce pseudocapacitance, accelerate the charge transfer, and facilitate the electrode/electrolyte interface reactions in BDCs. The incorporation of heteroatoms within BDCs usually alters the electron-accepting or donating characteristics, as well as the structures, thereby influencing the electrochemical performances. In general, O, N, P, and S elements...
inherited in biomass can form self-doped BDCs by controlled pyrolysis of biomass precursors.\textsuperscript{162–165} On the other hand, heteroatom-doped BDCs can be obtained by heteroatom-containing dopants.\textsuperscript{74}

Oxygen element exists in almost all BDCs consisting of oxygen-containing functional groups, such as hydroxy, carboxyl, and carbonyl, among others.\textsuperscript{166} This should enhance the surface wettability and increase ions accessible active surface areas, thereby introducing extra pseudocapacitance.\textsuperscript{167} In this view, BDC from elm seeds has shown an oxygen-rich porous structure with exceptional oxygen content (32.95 at.%), contributing to the pseudocapacitance and enhancing the surface wettability.\textsuperscript{157} BDC from camellia oleifera shell displays rich oxygen functional groups (C/O ratio of 1.66) after microwave-assisted carbonization and KOH activation, leading to excellent specific capacity when compared to others obtained through direct carbonization/KOH activation (high C/O ratio) in 2 M H\textsubscript{2}SO\textsubscript{4} (315 vs. 202 F g\textsuperscript{-1}) and 6 M KOH (251 vs. 214 F g\textsuperscript{-1}). This indicates that higher oxygen contents are beneficial for improving the specific capacitances of BDCs.\textsuperscript{111} On the other hand, the existence of oxygen functional groups is harmful to the capacitive performance of porous carbon in organic electrolyte. The previous results revealed that the removal of unstable surface oxygen of starch-based porous carbon further optimized the capacitive performance of ECs, delivering excellent suppressed self-discharge and leak current, and improved cycle life.\textsuperscript{168}

Nitrogen-doped BDCs with electron-donor properties usually improve the electrical conductivity, increase the electroactive sites, change the surface wettability and polarity through creating defects, and alter the valence electron orbital energy levels of carbon atoms.\textsuperscript{32} Four different forms of N-doping exist: pyridinic-N-oxide, graphitic-N, pyridinic-N, and pyrrolic-N\textsuperscript{169} (Figure 7). The positive charges pyridinic-N-oxide and graphitic-N are
beneficial for accelerating the electron transfer, while the negative charges of pyridinic-N and pyrrolic-N provide electroactive sites for pseudocapacitance to the specific capacity. Numerous biomass materials contain nitrogen element, including pine needle,164 juncus,46 chitosan,170 coronarium,148 pine pollen,171 bamboo fungus,172 spirulina platensis,173 and porcine bladders.174 These materials can be transformed into nitrogen self-doped BDCs by carbonization and activation without additional nitrogen dopants. However, most nitrogen self-doped BDCs suffer from low nitrogen contents, negatively affecting the electrochemical performances. Thus, the nitrogen sources, such as melamine,74,75,118,175 urea,96,176–178 dicyandiamide,83 ammonia,179 polyethyleneimine,176 and ammonium chloride,180 are widely used to increase the nitrogen content in BDCs. In this regard, BDC from peanut shell has shown nitrogen-doped porous structure with specific surface area of 1442 m²·g⁻¹ and nitrogen content of 3.2 at.%, leading to the higher specific capacitance of 216 F·g⁻¹ when compared to the material without nitrogen-doping (167 F·g⁻¹). This suggests that nitrogen doping could improve the specific capacity of BDCs.74 BDC from ginkgo leaf with nitrogen-doping and ultrahigh ratio of pyridinic-N/pyrrolic-N species (86.5%) has demonstrated better specific capacitance than BDCs from pristine ginkgo leaf and KOH-activated without nitrogen-doping.181 Besides, biomass itself contains both oxygen and nitrogen elements, which can facilitate the formation of O/N codoped BDCs, such as pigskin,182 perilla frutescens leaves,183 pine needle,127 and cattle bones.162

Similar to BDCs with oxygen- or nitrogen-doping, other elements-doping with sulfur, fluorine, and phosphorous can improve the electrochemical performances. Sulfur is an electron-rich reactive element that can be embedded into the carbon skeleton to serve as electroactive sites for redox reactions and modify its surface to improve the wettability.73 Furthermore, sulfur doping can increase the electrical conductivity because the lone pair of electrons in the p-orbital of sulfur atoms can overlap with the π-orbital of graphite sp² hybridization to generate electron delocalization along the conjugated carbon skeletons.184 On the other hand, fluorine functional groups could provide abundant C-F bonds and change the structure of BDCs, thereby affecting the electrochemical performances of BDCs. Note that fluorinated BDCs possess three kinds of C-F bonds: ionic, semi-ionic, and covalent bond induced by the different electronegativity values between carbon (2.5) and fluorine (4) atoms. In turn, these features generate variable BDCs performances.185 Like nitrogen, phosphorous shows the same number of valence electrons and chemical properties. However, the diameter of phosphorous is much larger than that of carbon, making phosphorous doping more beneficial for local structural distortion of the carbon framework and the creation of more active sites. By comparison, phosphorous has a lone pair electron, useful for inducing redox reactions and enhanced capacitance.186

The heteroatoms codoping also boosts the electrochemical properties of BDCs due to synergetic effect between the heteroatoms. Similar to the preparation of heteroatom-doped BDCs, heteroatoms codoped BDCs are often obtained by complex interactions between biomass precursors and heteroatom-containing dopants. This can also be achieved by pyrolysis of heteroatom-containing biomass, such as borassus flabellifer flower–derived S-doped carbon with thiourea as sulfur dopant,73 silkworm cocoon–derived N/P codoped porous carbon with phytic acid as phosphorous dopant,187 bacterial cellulose–derived N/P codoped nanofibers with (NH₄)₃PO₄ as phosphorous source,188 rice-husks-derived N/P codoped porous carbon with H₃PO₄ as phosphorous source,186 N/P-containing eleocharis dulcis,155 N/P-containing duckweeds,189 N/F-containing third-class red dates,190 N/O/S-containing natural biomass derivative kraft lignin,191 N/O/S-containing rape pollen,192 N/S/P-containing soybean flour,193 N/O/S-containing elm flower,194 and O/N/S-containing ant power.195

5.3 Graphitization degree

High graphitization degree leads to excellent electrical conductivity, as well as reduced internal resistance of electrode materials and accelerated charge transfer, thereby improving the power density and rate performance. High-temperature treatment is often used to obtain high graphitization degree carbon materials through increasing the sp² carbon atoms during the heating process.196,197 However, excessive heat treatment temperatures usually decline the specific surface area and pore volume due to pore collapse. This reduces the surface functional groups, seriously affecting the electrochemical performances of BDCs. Therefore, the balance should be kept among the graphitization degree, specific surface area, pore volume, and surface functional groups to obtain the optimal electrochemical performances.

For biomass, high graphitization degrees can easily be obtained by the hydrothermal. For example, extracts of prunus persica fruit can readily be converted into graphitization carbon by the hydrothermal carbonization of phytocconstituents (carbohydrates, organic acids, and dietary fiber) involving dehydration, polymerization, carbonization, aromatization, and passivation processes to yield a substantial graphitization degree (ID/IG = 0.65).198

The catalytic graphitization is another favorable strategy to improve the graphitization degree by catalysts.
during the activation process. Various graphitization catalysts, such as FeCl₃, K₂FeO₄, K₂CO₃, Fe(NO₃)₃, and NiCl₂ have so far been utilized to prepare BDCs with high graphitization degree. For instance, one-step carbonization and graphitization of bamboo char can be fulfilled by introducing the graphitization catalyst K₂FeO₄ at 800 °C, serving as an activator (K species) and graphitization catalyst (Fe species) to transform bamboo char into porous graphitization BDC from bamboo char. The resulting material shows the porous structure with specific surface area reaching up to 1732 m²·g⁻¹, as well as elevated electrical conductivity of 4.7 S·cm⁻¹. These values are much higher than those of bamboo char (4 m²·g⁻¹ and 0.2 S·cm⁻¹, respectively). Similarly, porous graphitization BDC from willow catkins can be obtained at 900 °C in the presence of K₄Fe(CN)₆, which does not only act as activator, but also as graphitization catalyst. The resulting BDC displays high graphitization degree (I_D/I_G ≈ 1.22) and superior specific surface area (1066.56 m²·g⁻¹), remarkably better than that of BDC from willow catkins without K₄Fe(CN)₆ (I_D/I_G ≈ 1.05 and 31.54 m²·g⁻¹, respectively). Coconut shells can easily be converted into porous graphene-like carbon sheets in the presence of K₂CO₃ at 900 °C. The obtained biocarbon shows specific surface area reaching up to 1506.19 m²·g⁻¹ and electrical conductivity as high as 32.14 S·cm⁻¹.

Additionally, the differences in structure and composition allow biomass to easily convert into high graphitization degree biocarbon through carbonization and activation processes. With regard to this, leaves of viburnum sargenti can transform into high graphitization degree (I_D/I_G = 0.73) porous carbon by low-temperature carbonization (500 °C) and KOH activation (600 °C).

5.4 | Defects
Defective carbon materials with topological defects, like vacancies and edges, might be used as active sites to adsorb more ions for improved capacitance and electrochemical performances. Therefore, carbon materials applied in ECs require moderate levels of structural defects for enhanced electroactive sites, as well as promoted ions adsorption, intercalation, and diffusion to yield enhanced specific capacitance values.

Numerous strategies have so far been employed to introduce defects in BDCs. Heteroatoms doping and subsequent removal is an effective way to form defective BDCs. In this respect, defective BDC from macadamia nut shells can be obtained by sequential hydrothermal treatment/carbonization/nitrogendoping/nitrogen removal strategy. After the removal of nitrogen atoms, defects are created in the carbon skeleton to serve as active sites for electrochemical reactions. For example, syzygium cumini leaves can convert into defective carbon nanosheets by NaHCO₃ activation at 850 °C to yield specific surface areas reaching up to 1184.4 m²·g⁻¹ and moderate levels of structural defects (I_D/I_G = 0.72), useful for more electroactive sites and elevated specific capacitance (222.26 F·g⁻¹). Mesopore-rich defective BDC from lotus leaves might be obtained through two-step activation approach. The first step consists of HNO₃ activation to introduce certain amounts of nitrogen in the carbon skeleton, while the second step relies on KOH activation to promote the removal of nitrogen atoms and generate some defects/pores. As a result, superior specific capacitance reaching up to 478 F·g⁻¹ is obtained with two-step activated BDC from lotus leaves, which is much higher than values delivered by one-step KNO₃ activated (173 F·g⁻¹) and nonactivated (40 F·g⁻¹). BDC with N/S/P codoped interconnected porous carbon nanosheets from soybean aerogel possesses nitrogen doping content of 5.83 at.%, which not only contribute the pseudocapacitance originates from pyridinic-N and pyrrolic-N, but also enhance the electrical conductivity. Meanwhile, this route also creates defects in the carbon, providing additional active sites and transmission channels for ions.

5.5 | Morphology
The morphology of carbon materials significantly impacts real applications. Natural biomass displays various morphologies, which can be converted into BDCs with different structures, including spherical, fibrous, tubular, sheet-like, and hierarchical skeleton structure.

5.5.1 | Spherical BDCs
Spherical BDCs are characterized by good mechanical strength and high bulk density, useful for increased volume energy density. Various methods have been utilized to prepare spherical BDCs, such as hydrothermal, pyrolysis, and self-assembly. Spherical BDCs can easily be obtained from biomass derivatives, such as sugar sources (glucose, saccharose, and xylole) by the hydrothermal process not directly from biomass. In this view, spherical BDC from xylose can be formed by the hydrothermal carbonization followed by sequential KOH-H₃PO₄ activation to obtain uniform spherical shaped porous structures with elevated specific surface areas up to 692 m²·g⁻¹ when compared to nonactivated BDCs (1.53 m²·g⁻¹, Figure 8A). For instance, spherical BDC from glucose and glucosamine with nitrogen-doped can be synthesized.
by aerosol-assisted process (Figure 8B), where the amino groups of glucosamine trigger self-assembly between glucosamine and glucose to form BDC with spherical morphology, suitable for high specific capacitance and long cycling stability. Porous hollow BDCs from soybean waste can be prepared by the hydrothermal carbonization followed by high-temperature treatment under different atmospheres. In Ar atmosphere (Figure 8C and D), BDCs with an average size of 398 ± 10 nm and average shell thickness of 40 nm are obtained after heat treatment. In mixed N<sub>2</sub> and H<sub>2</sub>O steam, high specific capacitance and low rate performances are noticed when compared to samples prepared by heat treatment in inert atmosphere (N<sub>2</sub> or Ar).

The appropriate biomass with microscale spherical structures, such as waste potato peel, might also be transformed into spherical BDCs by pyrolysis to preserve the morphology of the precursors. Various spores as carbon sources and self-templates could also be easily converted into spherical BDCs by facile carbonization and CO<sub>2</sub> activation route.

5.5.2 | Fibrous/tubular BDCs

Fibrous or tubular BDCs with unique morphology could serve as active electrode materials, conductive additives, and support substrates for other substances. Various methods have been employed to prepare fibrous BDCs, including pyrolysis, electrospinning, ultrasonication, and hydrothermal. Regarding this, fibrous BDC from hemp straw (Figure 9A) can be obtained by polyacrylonitrile assisted electrospinning followed by KOH activation. Fibrous BDC from sawdust might be acquired through pyrolysis of Fe<sup>3+</sup>-preloaded sawdust, which can effectively catalyze the growth of carbon nanofibers.

Compared to fibrous carbon, tubular carbon cannot only offer effective electron transmission paths, but also provide buffering space for fast ions diffusion or migration, thereby shortening the ions diffusion distances. Meanwhile, the rich pores in tubular carbon walls further increase the specific surface area for ions storage, as well as provide fast ions transport channels. Some biomass, such as enteromorpha prolifera, cotton, kapok fibers, and coconut fibers, possesses natural hollow tubular structure that can easily be converted into hollow tubular BDCs. For instance, BDCs from kapok fibers have tubular structures and N-containing sandwich-like walls, promoting ions transport. This, in turn, translates into excellent electrochemical performances with increased specific capacity when compared to BDC from cattail without tubular structure. As shown in Figure 9B, BDC from waste...
cottonier strobili fibers depict tubular structure with large accessible surface area and abundant micro-/mesopores, favoring ions storage and transfer, useful for high specific capacitance and excellent rate performance. Moreover, the structural advantages of fibrous or tubular BDCs can support pseudocapacitive materials to yield better electrochemical performances. In this respect, fibrous BDCs from cotton stalks as conducting matrix can prevent the aggregation of Ni-Al-layered double hydroxide nanosheets and enhance conductivity of the composite (Figure 9C). Consequently, the composite shows excellent electrochemical performances, prominently superior to those of Ni-Al-layered double hydroxide nanosheets.221 Hollow tubular BDCs from kapok fiber/NiO composite can be formed by the hydrothermal method with carbonization at 400 °C (Figure 9D). The kapok fiber serves as both carbon source and template for the growth of Ni(OH)2 on kapok fiber surface, during which Ni(OH)2 transforms into NiO accompanied by carbonization of kapok fiber. Meanwhile, the hollow tubular structure of kapok fiber is retained during the preparation process of hollow tubular BDCs/NiO composite.217 Similarly, BDCs from kapok-fibers-supported RuO2218 and BDCs from waste crab-shell-supported Co3O4 nanocrystals also exhibit enhanced electrochemical performances.222

5.5.3 Sheet-like BDCs

Carbon sheets with ultrathin thickness and large lateral size have unique layered structure, unusual physicochemical and electrical performance, as well as abundant accessible electrochemically active sites for ions adsorption/desorption.223 In particular, porous carbon sheets can provide additional specific surface areas, low resistance pathways, and shortened ions diffusion distances. Biomass can be changed into BDC sheets by template, thermal treatment, and carbonization-activation methods.51,78,224,225 For example, BDCs from larch wood sawdust with regular lamellar structure might be fabricated by liquefaction coupled with in-situ doping of tetraethoxysilane (silica-based template) followed by carbonization at 900 °C.226 BDCs from pomelo mesocarps with nitrogen-doped can be formed by one-step simultaneous carbonization (Figure 10A), CaCl2 activation, and urea nitrogen doping process to yield interconnected sheet-like structure.227 BDCs from peanut seed coats (Figure 10B) are constructed via triethanolamine intercalation followed by simultaneous thermal exfoliation and pyrolysis to form graphene-like structure with thickness of 4 nm.37 BDCs from coconut shells are synthesized by simultaneous activation/graphitization strategy. To this end, the activator
(ZnCl₂) and graphitization catalyst (FeCl₃) are simultaneously introduced into the coconut shells by coordination of the metal species with the functional groups in coconut shells under heat treatment. Meanwhile, Fe species in the skeleton of coconut shells generate carburized phase to form graphene-like structure during pyrolysis. Similarly, BDCs from coconut shells with graphene-like sheets with high specific surface area and excellent electrical conductivity (32.14 S·cm⁻¹) can be obtained by K₂CO₃ catalytic graphitization (Figure 10C), BDCs from walnut shells with pore walls composed of 1–2 carbon layers are prepared by carbonization and KOH activation to yield elevated electrical conductivities higher than YP-80F commercial porous carbon (720 vs. 93 S·m⁻¹), as well as superior specific capacitance and excellent rate performance. BDCs from eggplant with sheet-like structure can be prepared by one-step carbonization to generate excellent rate capability as the porous sheet-like structure provide short ions diffusion paths.

On the other hand, BDC sheets can act as a favorable substrate to support pseudocapacitive materials. Concerning this, BDCs from peach-gum-supported Ni(OH)₂ possess abundant active surface area (Figure 10D), beneficial for high specific capacity (than Ni(OH)₂), and superior power density. These improved features are linked to the carbon nanosheets, useful for enhancing the electrical conductivity and providing well-developed porosities for ions diffusion.

5.5.4 Hierarchical BDCs

As described in the pore structure, BDCs with hierarchical porous structure can boost the charge transfer, shorten the ions transport distance, and decrease the internal resistance due to the structural connectivity and high specific surface area. Therefore, hierarchical porous structures often show elevated ions and electrons conductivity, resulting in excellent electrochemical performances.

So far, numerous BDCs with hierarchical porous structure have been reported, such as coconut shell, cypress coats, rice husk, garlic skin, and waste shaddock endotheliums. For instance, BDCs from houttuynia with hierarchical porous illustrate high specific surface area (2090 m²·g⁻¹) and unique flower-like structure with well-distributed micro-/mesopores (Figure 11A), beneficial for enhanced energy density and cycling stability thanks to the inherent hierarchical porous structure. BDCs from rice straw with graphitic and interconnected pores have shown elevated specific surface area of 3333 m²·g⁻¹ with
abundant micro-/mesopores combined to high specific capacitance (400 F⋅g⁻¹), as well as favorable rate performance and cycling stability.²³¹ BDCs from pomelo peel depict hierarchical and interconnected honeycomb-like structure (Figure 11B), leading to better specific capacitance and excellent cycling stability.¹¹⁷ BDCs from wild jujube pit with hierarchical pores present interconnected micro-/meso-/macropores and advanced specific surface area of 2438 m²⋅g⁻¹ with pore volume of 1 cm³⋅g⁻¹ (Figure 11C), thereby delivering superior specific capacitance (398 F⋅g⁻¹ at 0.5 A⋅g⁻¹) and excellent rate performance (345 F⋅g⁻¹ at 20 A⋅g⁻¹).¹⁶⁶ BDCs from pomelo peel fiber display a hierarchical porous structure with a high oxygen content of 15.61 wt.% (Figure 11D), resulting in excellent electrochemical performance.²³² By analogy, hierarchical porous BDCs from mixed wastes of coconut shell and sewage sludge show high specific surface area of 3003 m²⋅g⁻¹, large pore volume of 2.04 cm³⋅g⁻¹ and appropriate porosity, resulting in excellent electrochemical performances.¹²²

In summary, BDCs with different morphologies can lead to different electrochemical performances. Therefore, the development of BDCs with controllable morphologies is highly desirable for improving the electrochemical properties. In this regard, BDCs from aloe peel undergoing morphology transformation from spherical to honeycomb-like structure have shown an increase in specific surface area from 13 to 1286 m²⋅g⁻¹, as well as improvement in the specific capacitance.²³³ BDCs from pine barks with tunable morphologies, such as honeycomb-like porous carbons, uniform carbon nanosheets, and irregular-shaped carbon microparticles, deliver different specific capacitances and rate performances attributed to the differences in morphologies and structural characteristics.²³⁴

5.6 | BDCs composites for ECs

5.6.1 | BDCs with graphene or carbon nanotubes composites

Graphene and carbon nanofibers/nanotubes often exhibit excellent physicochemical properties in terms of large specific surface area, high electrical conductivity, and good flexibility. Therefore, the graphene or carbon nanofibers/nanotubes in BDCs can improve the electrochemical capacitance values.

Graphene could provide sufficient space for loading active materials due to its 2D structure, leading to large specific surface areas and elevated electrical conductivities. For example, the combination of BDCs from coffee grounds with reduced graphene oxide (PAC-rGO) leads to enhanced electrochemical performances. The reason has to do with the incorporation of reduced
graphene oxide in BDCs from coffee grounds, which form 3D interconnected conductive network among BDCs from coffee grounds particles (Figure 12A–C). This, in turn, offers fast ions diffusion and charge transfer, as well as increases the accessible surface area of ions, and prominently improves the specific capacity of BDCs from coffee grounds. The excellent cycling stability and high specific capacitance of 324 F·g⁻¹ are recorded, far better than values obtained without reduced graphene oxide (93 F·g⁻¹, Figures 12D–E). On the other hand, graphene could effectively restrict the volume expansion of bulk BDCs during cycling. For example, the graphene in BDCs from coconut shell enhances the electrochemical performances because the graphene might support particles and limits the volume expansion during the charge/discharge processes (Figures 13A and B). Briefly, composite electrodes...
with 3% graphene deliver expansion rates of 11%, smaller than the 3% ketjen black (KB, conductive agent) with 27% expansion rate. The composites exhibit better capacitance performance (173 F g\(^{-1}\)) than that with KB addition (153 F g\(^{-1}\)). Therefore, the graphene enhances the electrochemical performances (Figures 13C–J).\(^{236}\) Other BDCs from cornstalk-core\(^{237}\) reed straw,\(^{238}\) milk protein\(^{239}\) and cellulose paper\(^{240}\) coupled with graphene also display good electrochemical performances.

The carbon nanofibers/nanotubes should also improve the BDCs electrochemical performances. For instance, BDCs from sawdust transform into nanofibers/mesoporous carbon composite by fast pyrolysis of FeCl\(_3\)-preloading sawdust. FeCl\(_3\) can promote the decomposition of cellulose and hemicellulose into small molecular hydrocarbons, which can deposit on the formed Fe species to grow carbon nanofibers similar to chemical vapor deposition process. The presence of both Fe and Cl species could effectively catalyze the growth of carbon nanofibers and form magnetic nanofibers/mesoporous carbon composites. Higher specific capacitance values reaching 89 F g\(^{-1}\) are obtained with FeCl\(_3\)-preloading composites when compared to composites without carbon nanofiber by fast pyrolysis Fe(NO\(_3\))\(_3\) or Fe\(_2\)(SO\(_4\))\(_3\)-preloading sawdust (19 or 21 F g\(^{-1}\), respectively). Hence, the presence of nanofiber structures might effectively facilitate the diffusion of ions and the transport of electrons among carbon particles.\(^{216}\)

Compared to carbon nanofibers, carbon nanotubes possess higher specific surface areas and more diffusion channels for ions. Carbon nanotubes often affect the morphological and textural properties of BDCs during the hydrothermal synthesis, thereby influencing the final electrochemical characteristics. For example, the carbon nanotubes impact the morphology and electrochemical performances of BDCs obtained by facile hydrothermal processes of glucose. The addition of different contents of carbon nanotubes results in various morphologies and physicochemical properties of BDCs from glucose. Notably, the morphology of BDCs can evolve from spherical to platelet-like particles with slit-shaped pores (~400 nm). The obtained composite with 2 wt.% carbon nanotubes followed by KOH chemical activation yields slightly higher specific capacitance of 220 F g\(^{-1}\) than that of without carbon nanotubes (218 F g\(^{-1}\)) at 0.2 A g\(^{-1}\). In addition, the former exhibit better capacitance retention such as 22% capacitance delay at 20 A g\(^{-1}\) than the latter such as more than 50% capacitance delay even at 5 A g\(^{-1}\). In summary, carbon nanotubes improve the electrochemical properties of BDCs from glucose greatly when it used in ECs.\(^{35}\)

The increase in specific surface area of carbon material without significant reduction in conductivity is effective strategy to improve the electrochemical performances. The regular tubular tracheids structures and conductive channels of wood are conducive to obtain BDCs with high conductivity and large specific surface area through rational structural design. For instance, carbon nanotubes/BDCs from wood composites with special structures can be obtained through chemical vapor deposition (Figure 14A). Tracheids size of 20–30 μm and aligned carbon nanotubes arrays with 3 μm in height are grown on curved inner walls of wood tracheids (Figures 14B–E). Such architectures effectively improve the space utilization of BDCs from wood and increase the electrochemical performances. Carbon nanotubes grown on inner wall of tracheids of BDCs from wood lead to improved specific surface area from 365.5 to 537.9 m\(^2\) g\(^{-1}\), thereby improving the specific capacity of BDCs from wood up to 215.3 F g\(^{-1}\) (or 76.5 F cm\(^{-3}\)). The resulting composite electrodes also show excellent flexibility when assembled into flexible self-supporting multunit ECs.\(^{241}\) Overall, the full use of natural tubular tracheids structures of woody biomass combined with the rational structural design such as carbon nanotubes is beneficial for improving the electrochemical performances.

### 5.6.2 BDCs with pseudocapacitive materials composites

Compared to carbon materials, pseudocapacitive materials based on typical transition metal oxides or hydroxides, such as MnO\(_2\),\(^{242–244}\) Fe\(_2\)O\(_3\),\(^{245}\) NiO,\(^{217}\) Co\(_3\)O\(_4\),\(^{222,246}\) and Ni(OH)\(_2\),\(^{230}\) possess higher capacity and superior redox activity. The redox reactivity of the materials mainly originates from the multivalent characters of transition metals. However, the lower specific surface area, inferior intrinsic electrical conductivity, and severe structural collapse under high current density cycling lead to slow down the charge transfer process and unsatisfactory cycling stability.

BDCs can offer abundant space to deposit transition metal oxides or hydroxides, as well as reduce agglomeration. Meanwhile, the superior electrical conductivity of BDCs could accelerate the charge transfer, thereby improving the rate performance and power density. Besides, the excellent structural stability and appropriate porous architecture of BDCs could buffer the volume expansion induced by transition metal oxides or hydroxides during the charge/discharge.\(^{243,247}\) Therefore, the combination of transition metal oxides or hydroxides with BDCs could improve the electrochemical performances.

MnO\(_2\) is representative pseudocapacitive material with high theoretical specific capacity of 1370 F g\(^{-1}\).\(^{247}\) The charge storage mechanism of MnO\(_2\) mainly originates from surface reaction processes during adsorption/desorption and insertion/extraction of alkali cations and...
proton (Equation (6)).

$$\text{MnO}_2 + M^+ + e^- \rightarrow \text{MnOOM (M = H, Li, Na, or K).} \quad (6)$$

However, the low electrical conductivity and inferior cycling stability of MnO$_2$ dramatically decline the electrochemical capacitive properties. To overcome these drawbacks, MnO$_2$ is often composited with BDCs to enhance the electrochemical performances. In this regard, BDC from faidherbia albida fruit shell combined with MnO$_2$ leads to the formation of MnO$_2$ with nanowire-like structure grafted on BDC and excellent electrochemical performances. In this process, BDC is first obtained by hydrothermal carbonization and H$_3$PO$_4$ activation to serve as supporting framework to grow MnO$_2$ through hydrothermal synthesis. The synergistic effects of BDC and MnO$_2$ yield MnO$_2$/BDC with excellent flexibility and elevated specific capacitance values of 426 and 202.5 F·g$^{-1}$ in 1 M KOH and 1 M Na$_2$SO$_4$, respectively, when compared to BDC (333 and 118 F·g$^{-1}$, respectively). Similarly, BDC from wheat bran with MnO$_2$ nanosheets delivers substantial specific capacitance and rate performance. The MnO$_2$@BDC composite can be prepared through two-step process. In detail, BDC is first obtained by one-step pyrolysis carbonization using NaCl/ZnCl$_2$ as activator, and MnO$_2$-loaded BDC is formed by the hydrothermal process. The MnO$_2$ can improve the specific capacity of BDC, with MnO$_2$@BDC revealing high specific capacitance of 258 F·g$^{-1}$ at 1 A·g$^{-1}$ and excellent rate performance of 235 F·g$^{-1}$ at 20 A·g$^{-1}$. These values are superior to BDC.

Other pseudocapacitive materials such as Fe$_2$O$_3$, Co$_3$O$_4$, NiO, and Ni(OH)$_2$ can also combine with BDCs to enhance the capacitance performances. More importantly, the BDCs with special structures might be used as substrates to support nanostructured NiO and take full advantage of inborn structural of BDCs. In this respect, kapok fiber has natural hollow tubular structure that can serve as substrate for supporting NiO. Hence, BDC from hollow tubular kapok fiber coupled with NiO nanosheets (BDC/NiO) is prepared by the hydrothermal and carbonization processes. The unique microtubular structure combined with the synergetic effects between BDC and NiO forms BDC/NiO with substantial specific capacitance and excellent cycling stability. BDC from wheat straw is combined with Fe$_2$O$_3$ ultrathin film though two-step process. The first step consists of direct pyrolysis of wheat straw using KOH as an activator to obtain BDC. The second step deals with annealing of the composite of BDC and precursor of Fe$_2$O$_3$ (Figure 15A). The hierarchical porous structure and synergetic effect between BDC and Fe$_2$O$_3$ yields BDC/Fe$_2$O$_3$ with higher specific capacitance than BDC and Fe$_2$O$_3$, combined with and excellent cycle performance (Figure 15B–D).

Composites containing different BDCs and transition metal oxides or hydroxides, such as BDC from terminalia chebula fruit/Co$_3$O$_4$, BDC from peach gum/Ni(OH)$_2$, BDC from persimmon fruit/Fe$_2$O$_3$, BDC from shiitake mushroom/NiCo$_2$O$_4$, BDC from kapok fiber carbon aerogel/RuO$_2$, and BDC from sugar-cane bagasse/MnO$_2$, have been used as electrode materials of ECs.

Other popular pseudocapacitive materials include conducting polymers, such as polyaniline (PANI), polypyrrole (PPy), and polythiophene (PTP). These materials have been considered to improve capacitance performances. However, conducting polymers have poor rate performance and inferior cycling stability ascribed to the limited ions diffusion, volume shrinkage, and expansion during the charge/discharge cycling. Among conducting polymers, PANI is commonly used as additive in BDCs from various biomass, including seaweed, kenaf, wheat flour,
enteromorpha prolifera, celery, willow catkin, and watermelon rind. For example, BDC from dandelion fluff/polyaniline composite (ACTBs/PANI) is obtained by two-step process consisting of first transforming hollow tubular dandelion fluff into BDC through carbonization and KOH activation, followed by coating of ACTBs with PANI by in-situ chemical polymerization (Figure 16A). The porous hollow tube structure of ACTBs and the introduction of pseudocapacitive PANI form ACTBs/PANI composite with excellent capacitance retention and cycling stability (Figure 16B–G). BDC from chitosan/polyaniline composite can be obtained via self-assembly and ice template followed by polymerization process to yield largely enhanced electrochemical performances. The hierarchical porous structure of BDC from chitosan does not only allow the infiltration
and uniform coating of PANI in the inner framework, but also facilitates the diffusion of the ions during the charge/discharge cycling. Meanwhile, the synergetic effect of the double-layer capacitance of BDC from chitosan and pseudocapacitive of PANI are useful features for improving the capacitance performances. Hence, BDC from chitosan/PANI exhibits higher specific capacitance when compared to BDC from chitosan.266

Like polyaniline, polypyrrole can also improve the electrochemical performances of BDCs. Numerous reports indicate the improved electrochemical performances of polypyrrole with BDCs from bacterial cellulose nanofibers,267 cattail,268 bamboo cellulose nanofibers,269 and durian rind.270 In sum, the pseudocapacitive materials can improve the electrochemical performance of BDCs due to the synergetic effects.

6 | CHALLENGES AND OUTLOOKS

BDCs are promising electrode materials in energy storage fields due to abundant natural resources, low cost, environmental friendliness, sustainability, renewability, adjustable pore structure, and excellent physicochemical stability. The rational design and preparation of BDCs with adjustment of specific surface area, pore structure, and surface chemistry are important features for the application in ECs. The heteroatoms self-doping, graphitization degree, defects, and morphologies also seriously affect the properties of BDCs, thereby should be considered in future development and progress of BDCs. BDCs compositing with other materials, such as graphene, carbon nanofibers/nanotubes, transition metal oxides or hydroxides, and conducting polymers, could be taken into account to further modify the electrochemical performance. The potential challenges of BDCs are summarized below:

1. The diversity of biomass requires further research of BDCs originating from different sources to gain better controllability. BDCs obtained from mixtures of different biomass precursors also need further studies. Meanwhile, the sources and preparation ways should be expanded. For instance, bacteria adsorbing metal ions can directly be carbonized and oxidized to yield composite structures with potential applications.

2. The precise regulation of the pore structure of BDCs needs further exploration, such as BDC from coconut shells. Other BDCs already in use, more precise regulation should be set up.

3. Novel preparation methods and mass production of BDCs from current promising laboratory data are necessary for industrial upscaling. Meanwhile, the economic benefits and investment costs should be considered, especially for large-scale utilization of economically valuable biomass.

In addition to the above challenges, extending the applications of BDCs is worth considering. The diversity in sources, species, structures, and compositions of biomass endows the composites with wide ranges of applications (Figure 17), such as in seawater desalination,271 lead-carbon batteries,272,273 lithium-ion batteries,274,275 lithium–sulfur batteries,276–278 lithium metal batteries,279,280 sodium-ion batteries,279,281 oil/water separation,87 and catalysts (or catalyst supports).282,283 For example, BDCs with high specific surface area with appropriate pore size and pore size distributions are beneficial for use in seawater desalination, water treatment, and oil/water separation. Similarly, these materials also act as catalysts or substrates supporting metal-based catalysts for electrocatalysis applications. Meanwhile, BDCs with heteroatoms modification might possess altered surface characteristics, such as defects, active sites, and interlayer spacing. These features are conducive to intercalation/deintercalation of Li+/Na+ in lithium/sodium ions batteries/capacitors. Numerous recent advances in BDCs for advanced energy storage techniques and devices have also been reviewed in some other articles.284,285 Furthermore, besides EDLCs, other types of ECs, such as pseudocapacitors or asymmetric capacitors, should have different requirements for BDCs electrode materials and have to be considered.

In summary, the various advantages of BDCs are promising for a wider range of applications but more efforts are required for future advanced prospects.
CONFLICT OF INTEREST
The authors declare no conflict of interest.
[Correction added on 29 June 2021, after first online publication: Conflict of Interest section has been added.]

ACKNOWLEDGMENTS
This study was financially supported by National Natural Science Foundation of China (52062012, 51525206, 520105010, and 21603048) and Key Science & Technology Project of Hainan Province (ZDYF2020028).

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**How to cite this article:** Luo X, Chen S, Hu T, Chen Y, Li F. Renewable biomass-derived carbons for electrochemical capacitor applications. *SusMat*. 2021;1:211–240. https://doi.org/10.1002/sus2.8