



# Recent progress in post-modified biochar-based material for supercapacitor applications: a review

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Received: 20 February 2025 / Accepted: 16 October 2025  
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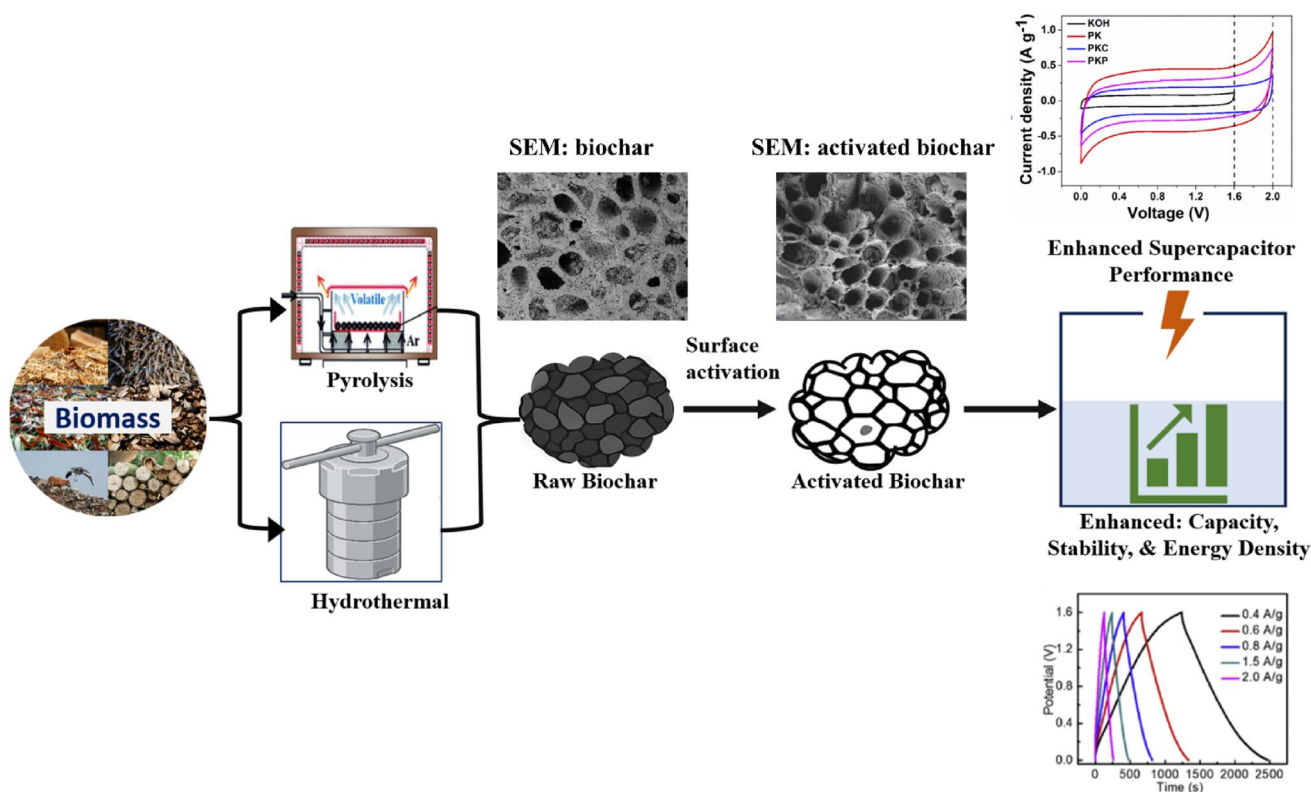
## Abstract

The escalating demand for efficient and sustainable energy storage solutions has spotlighted post-modified biochar materials as promising candidates for supercapacitor electrodes due to their high power density, rapid charge/discharge rates, and long-term stability. This review provides a comprehensive analysis of recent advancements in the synthesis, activation, and functionalization of biochar for supercapacitor applications. Various biomass sources, including agricultural and industrial wastes, have been pyrolysed or hydrothermally carbonised and further activated using agents such as KOH, NaOH, ZnCl<sub>2</sub>, and H<sub>3</sub>PO<sub>4</sub>, achieving specific surface areas (SSA) as high as 3577 m<sup>2</sup>/g and pore volumes up to 2.3 cm<sup>3</sup>/g. The electrochemical performance is significantly enhanced through heteroatom doping (N, O, S, P) and metal oxide composite formation, leading to specific capacitances ranging from 252 F/g to 550 F/g and energy densities up to 45.69 Wh/kg. Further, surface modification improves wettability and electron transport while mesopore/hierarchical structures facilitate ion diffusion. The nitrogen-doped biochar demonstrated a specific capacitance of 420 F g<sup>-1</sup> at 1 A g<sup>-1</sup>.m, whereas KOH-activated walnut shell-derived biochar exhibited 3577 m<sup>2</sup>/g SSA and 81% capacitance retention over 5000 cycles. Also, surface oxidation techniques have improved wettability and charge transfer, leading to excellent long-term cycling stability, with capacitance retention above 95% after 10,000 cycles. Owing to increased attention towards eco-friendly, viable, and scalable energy solutions, this article presents a thorough overview of the advanced techniques to treat biochar as supercapacitors. Challenges such as scalability, performance, and cost-effectiveness are presented, and a discussion of the future outlook for integrating biochar for sustainable energy storage is provided.

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## Graphical abstract



**Keywords** Activation methods · Biomass · Biochar · Electrode materials · Supercapacitors · Thermochemical techniques

## Introduction

The rapid depletion of fossil fuels to meet the global energy needs has prompted policymakers and researchers to explore alternative, renewable, sustainable, and cleaner sources of energy. In this quest, several approaches have been suggested to address these issues [1, 2]. Harnessing renewable energy sources is considered viable and of greater importance due to their ability to provide clean and sustainable energy for future generations [3, 4]. Researchers have successfully utilised these renewable energy sources, such as solar, tidal, wind, and hydropower, to meet the energy demands [5, 6]. These renewable energy sources are significantly affected by climatic changes, wind patterns, and seasonal variations, thereby posing challenges in ensuring an uninterrupted energy supply. Thus, an uninterrupted power supply is expected for all practical applications using effective energy storage technologies [7]. Therefore, designing and developing eco-friendly and efficient energy conversion and storage devices and technologies is imperative. Batteries and supercapacitors are the energy storage systems shown to have enormous potential [8]. Researchers have shown that

supercapacitors have more energy density, long life cycles, stability, fast charging and discharging times, and power density than regular capacitors. It also offers additional benefits, such as a broad working temperature range and high electrical conductivity [9]. Supercapacitors can be divided into pseudo-capacitors (PCs) and electrochemical double-layer capacitors (EDLCs), depending on their function. These capacitors primarily use electrostatic force to store charges on electrodes to provide energy whenever required [10]. Supercapacitors can perform a steady charging cycle for a long gadget life and have optimal power values recognition due to the electrochemical double-layer phenomenon. Reversible oxidation-reduction serves as the foundation for pseudocapacitors operating principle. Its electrode materials typically consist of transition metal-based oxides, alloys, conductive polymers, and composite materials, which can offer high specific capacitance. Therefore, heteroatoms and metallic atoms are often added to the electrode material to increase its pseudo-capacitance capabilities [11]. Pseudocapacitors, which rely on cyclic oxidation-reduction processes, have poor cycle stability and inferior life span, thus limiting their potential applications [12]. A prospective

solution to this problem is to harness biochar in supercapacitor applications. Biochar is a valuable carbon-rich material made from a sequence of thermochemical processes from biomass [13]. The carbon materials obtained from biomass have been realised as viable materials for supercapacitors owing to their desirable physicochemical properties such as high surface area, porosity, functional groups, conductivity, stable structure, and morphology for ion mobility [14, 15]. Numerous studies have revealed that the pyrolysis and other processes used to convert biomass into biochar are carbon-negative. The whole carbon cycle of a traditional biomass fixed-bed pyrolysis facility in China was examined by Yang et al. [16]. The intensity of carbon gas emissions was found to be  $1.55 \times 10^{-02}$  kg CO<sub>2</sub>-eq/MJ. However, if 41.02% of the biochar is put back into the field, the net emissions of greenhouse gases will be zero, indicating that the entire carbon cycle can be renewable. The significant amount of organic matter and carbon components present in biochar can significantly reduce atmospheric carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>), nitrous oxides (NO<sub>x</sub>), and fluorinated gases [17, 18]. In addition to decreased dependence on fossil fuels and reduced greenhouse gas emissions, the preparation of biochar also results in bio-oil production [19]. Because biomass is a abundant resource, it can be regarded as a viable source to produce fuels that can be used as a substitute for fossil fuels. Also, carbon materials produced from biomass are an affordable and environmentally friendly process [20]. The commonly used, carbon-based materials to produce supercapacitors are shown in Fig. 1 (a), which illustrates various biomass sources used for biochar production in supercapacitor applications. These include mulberry, corn stalk, coffee grounds, algae, fungus, organic waste, food waste, and bagasse. These sustainable and renewable feedstocks highlight the eco-friendly approach to developing high-performance energy storage materials for supercapacitors through the conversion of organic waste into functional carbon-based materials.

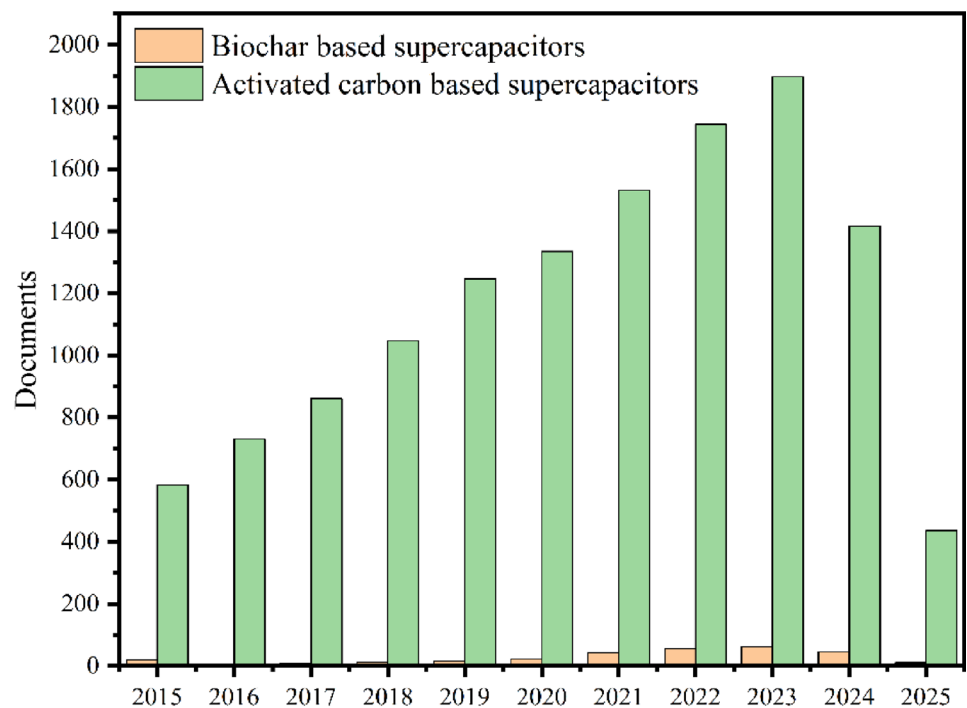
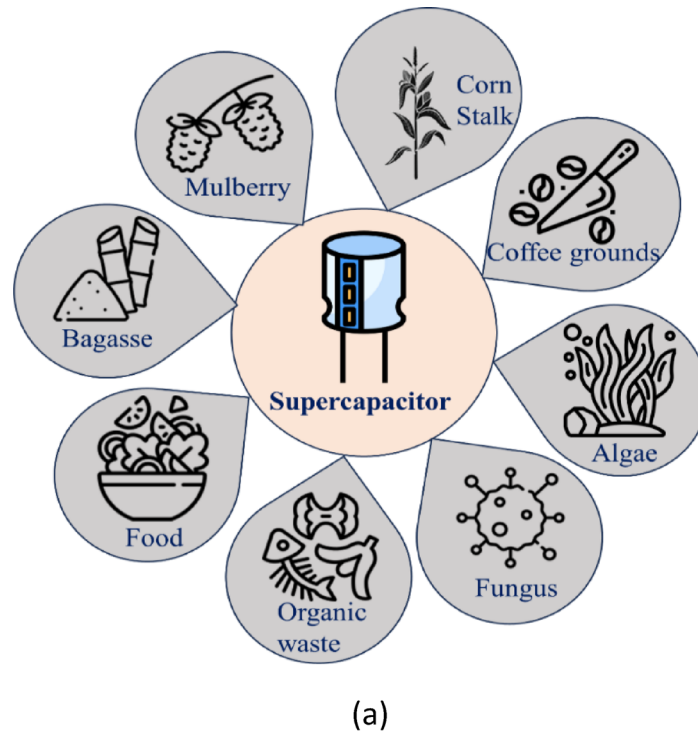
By tuning the preparation techniques, biochar can be engineered for the promotion of various functional groups, which in turn ensures a spectrum of physical and chemical characteristics [21]. Biochar with various morphologies, including fibrous materials, nanospheres, sheet-like structures, and honeycombs, can be created from different biomass owing to their specific compositions [22]. Biochar with post-modification has demonstrated a high specific surface area (SSA), hydrophilic functional groups, hierarchical pore structure, and superior conductivity [23]. An extensive literature review has indicated that there are few research articles discussing biochar for supercapacitors derived from lignocellulosic biomass. These articles focus only on agriculture-based and waste residue and do not provide a comprehensive explanation of the spectrum

of biochar-based materials. This review article commences with a concise analysis of the literature regarding the various applications of biochar or post-modified biochar. This study also encompasses the synthesis and activation methods of porous carbons produced from biomass for application in supercapacitors, as well as the critical factors of biochar that influence its performance as an electrode in such devices. Subsequently, this article introduces supercapacitors as devices for electrochemical energy storage. Additionally, the discussion encompasses the pivotal role of carbon in supercapacitors, mainly the carbon derived from activated biochar. Moreover, the article explores an array of techniques implemented to fabricate biochar possessing desirable attributes, including a high surface area, mesopore content, hierarchical pore structure, and high energy density. It is noteworthy to mention that this article also endeavours to examine the utilisation of green electrolytes and the recycling of supercapacitors, thus highlighting their significance in the pursuit of a sustainable environment. Recent research endeavours emphasise the role of biomass-derived materials and carbon-based materials in supercapacitors and hybrid capacitors in improving the energy storage abilities. The progress in this field includes engineered biochar, novel anode materials, and hybrid capacitors. The practical application of biomass-based supercapacitors is still facing challenges due to scalability, electrochemical stability, and the structure-property relationship [24–30]. The Biochar-based supercapacitors and activated carbon-based supercapacitors were searched on the Scopus database, and documents were presented in Fig. 1(b). Figure 1(b) illustrates a clear trend in research publications from 2015 to 2025 (10-year gap), showing a dominant focus on activated carbon-based supercapacitors, which have seen a steady rise in documents, peaking in 2023. In contrast, biochar-based supercapacitor research, although significantly lower in publication volume, has shown a gradual increase, especially post-2020. This indicates a growing scientific interest in biochar as a sustainable and low-cost alternative material. While activated carbon remains the mainstream choice, biochar's emergence reflects its potential in green energy storage technologies, particularly due to its renewable nature, carbon negativity, and alignment with circular economy principles. Lastly, with a few exceptions, this review paper is dedicated to examining and evaluating articles published within the last 10 years (except a few).

## Biochar and hydrochar

Harnessing biochar for applications like renewable energy, carbon sequestration, and soil development is a relatively new concept. Biochar is a solid carbon material obtained

**Fig. 1** (a) Biochar production from viable biomass feedstocks for supercapacitors (b) Documents related to biochar-based supercapacitors and supercapacitors are available in the Scopus database



(b)

through the thermochemical transformation of biomass feedstock in an atmosphere with limited oxygen [22]. It improves the soil nutrients and water retention and are helpful in preventing soil fertility because of climate changes. Other applications include water and wastewater remediation,

adsorbents, and carbon electrode materials in supercapacitors for sustainable energy conversion and storage [31]. On the other hand, hydrochar is a substance that resembles biochar in some ways but is produced under entirely different pre-treatment conditions. Biochar is produced via a

dry carbonization method like pyrolysis, unlike hydrochar, which makes a slurry by hydrothermal carbonization (HTC) using water as a medium. The chars produced from these two methods have a distinct physical and chemical makeup [19]. Biochar is used as a solid fuel for the combustion process or as a raw material to prepare electrodes for electrochemical energy conversion and storage cells like fuel cells, supercapacitors, electrolysers and ion-batteries; hydrochar has a variety of uses in the energy industry [32]. Unactivated biochar has a limited surface area, therefore exhibiting its limitation in being used as an adsorbent. The activation process enhances the adsorption efficiency and energy storage capacity of the carbon material, i.e., the biochar. Though the presence of functional groups on biochar enhances adsorption efficacy, the availability of fewer functional groups, such as hydroxyl ( $-\text{OH}$ ) and carbonyl ( $\text{C}=\text{O}$ ) groups, would not contribute significantly to adsorption [33]. Researchers in the last decade have revealed that additional functional groups on the biochar have enhanced the ability to adsorb dyes and heavy metals. Here, acidic oxy-functional groups on the biochar augment the electrostatic interaction between negatively charged adsorbate molecules such as heavy metals and cationic dyes [34]. The low surface area and porosity of hydrochar resulting from forming hydrocarbons on its surface restrict its usage as a contaminant adsorbent and catalyst base. Physical or chemical activation, surface functionalization and other alterations can improve hydrochar's porous structure and physicochemical characteristics. Hydrochar is slightly more acidic than biochar because it has more oxygenated functional groups. Due to the inorganics being removed during HTC in the water medium, hydrochar has an acidic pH [35]. The hydrochar can be functionalized at an acidic pH by scattering or sulfonating metals. For catalytic uses, activated carbon follows physical or chemical activation. The simplest way to change hydrochar characteristics is by functionalization, which involves subjecting the hydrochar surface to specific functional groups. It may be modified to function as an acid catalyst in procedures like the manufacture of biodiesel by adding acidic functional groups such as sulphonyl ( $-\text{SO}_3\text{H}$ ) or carboxyl ( $-\text{COOH}$ ) to the hydrochar matrix [36]. Biochar can be derived from diverse sources of waste, including plant/forest waste, waste from biological industrial processes, municipal solid waste, algae, and livestock [37]. When employing biochar as a material for supercapacitors, it is imperative to take into account the availability and cost of these waste materials. Agricultural waste, for instance, has attracted attention due to its sustainability and affordability [38]. Furthermore, the utilisation of waste materials such as Sapindus peels and oyster shell waste can further diminish the expense of biochar production [39]. The creation of biochar from biomass residues, such as *Tenebrio molitor* faeces, also presents a

cost-effective and environmentally friendly approach [40]. It has been reported that biochar derived from coconut shells costs approximately \$5-\$40 per kg [41]. Therefore, the consideration of waste material availability and cost is of utmost importance when employing biochar as a material for supercapacitors.

### Biochar properties affecting the performance of supercapacitor

The particular structure of the precursor is often carried over into the biochar material, such as fibre structure [42], layered structure [43], and carbon skeleton structure [44], which meets the present needs of biochar electrodes for supercapacitor applications. In the electrochemical double-layer energy storage process, a supercapacitor's specific capacitance may be successfully increased by increasing the number of active sites in the electrode material [45]. The specific capacitance offered by a supercapacitor should, theoretically, be proportional to the SSA of the carbon material, which increases with increasing SSA. Increased SSA is a successful strategy for boosting the number of active sites in several investigations [46]. However, it is reported that an increase in SSA does not linearly increase the specific capacitance of the biochar electrode materials. Small pore size micro-pores significantly contribute to the SSA because larger electrolyte ions cannot fit through the tiny holes. Numerous studies have demonstrated that materials with a high fraction of micro-pores do not function electrochemically as well as they should because they do not allow for the efficient movement of ions, particularly those from organic electrolytes, between electrode materials [47]. The rate of transmission of electrolyte ions will be markedly accelerated if the pore size in the electrode materials and the size of the electrolyte ions are compatible. The material's capacitance performance needs to be improved in order to use its SSA better. Hence, the pores must have the proper size and diameter [48]. Significant types of surface chemical alteration include inserting metal or non-metal heteroatoms and encouraging the synthesis of functional groups with oxygen on the surface. The surface-related properties, such as wettability and pseudo-capacitance of electrode materials, can be tailored by modifying the surface functional groups ( $-\text{OH}$ ,  $-\text{COOH}$ ) and heteroatoms (N, O, and P) doping, which is expected to impact the electrochemical efficiency [49]. In addition to influencing conductivity and wettability, surface heteroatoms can induce pseudo-capacitance through redox processes [42]. Nevertheless, functional groups and heteroatoms may exacerbate the self-discharge phenomena, decrease stability, and cause the supercapacitor's leakage current. The electrochemical performance

of supercapacitors is significantly affected by the internal resistance of the biochar electrode materials [50]. A significant amount of disordered carbon structures will emerge during the synthesis of biochar materials through the impact of specific activators, which is beneficial to the SSA but detrimental to conductivity [50]. The conductivity and degree of graphitization increase with reaction temperature, but impede the pore structure [51]. It is essential to arrive at reasonable decisions when processing biomass for specific applications.

## Methods for the preparation of biochar

The conversion techniques are dictated by the kind of biomass and the intended application of the final product [22]. This section details the numerous thermo-chemical conversion techniques for producing a variety of materials with factual value and their properties [52]. The torrefaction of biomass, a mild form of pyrolysis at 200–300 °C creates biochar, whereas hydrothermal carbonization (HTC), which occurs at around 200–350 °C, creates hydrochar as a slurry in the waste [53, 54]. The most popular processes for producing porous carbon are simultaneous thermo-chemical transformation and activation of both biochar and hydrochar materials in a single reactor.

### Pyrolysis

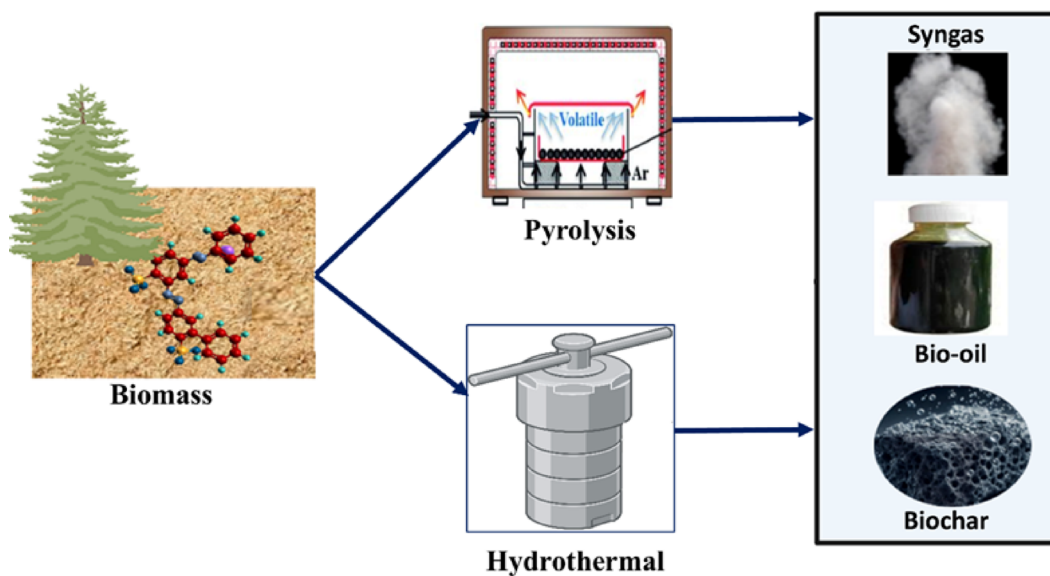
Pyrolysis is a thermochemical process that may break down biomass to produce charcoal in oxygen-limited or oxygen-free environments [55]. Pyrolysis has evolved into a versatile process that can create various end products as science and technology progress. This process produces solid, liquid, and gaseous products and can be classified as slow, moderate, or rapid based on residence duration and heating rate [56]. Commonly, slow pyrolysis is employed to create solid biochar or charcoal products. Typically, slow pyrolysis yields 25–30% liquid, 30–35% syngas, and 30–35% solid compounds [54]. A detailed study of the pyrolysis process is discussed in our earlier studies [22]. The schematic representation of the biochar preparation using pyrolysis methods is presented in Fig. 2 (a). Biomass, derived from organic sources such as plants, undergoes pyrolysis in the absence of oxygen at elevated temperatures (400–900 °C) at a lower heating rate (< 50 °C/min), resulting in the formation of syngas, bio-oil, and solid biochar. These processes enhance the value of biomass by converting it into functional materials for energy storage and other applications. Among the products, biochar is particularly significant for its application as supercapacitor electrode owed to its porous structure.

### Hydrothermal carbonisation

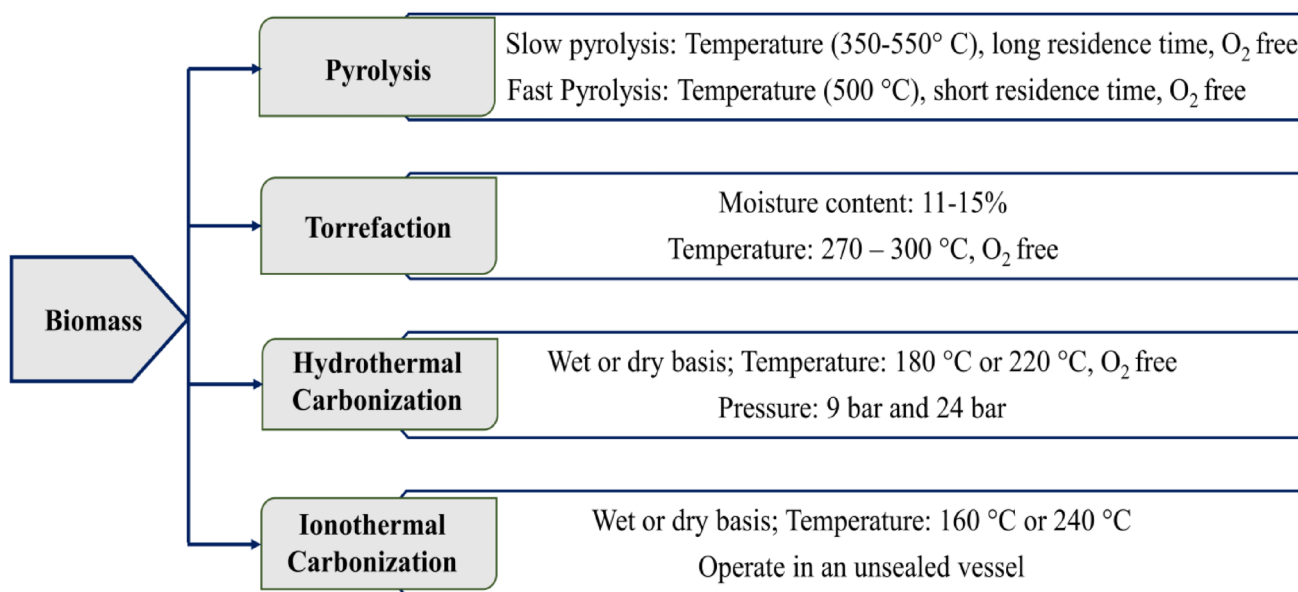
Hydrothermal carbonisation is a thermo-chemical process typically carried out in an aqueous solution at 150–350 °C [13]. This technique produces polycyclic aromatic hydrocarbons with oxy-functional groups and furans, in contrast to carbon materials created by the pyrolysis technique [19]. The hydrothermal treatment of biomass comprises a series of five steps: hydrolysis, dehydration, decarboxylation, polymerization, and aromatic cyclisation [19]. Researchers witnessed that biochar has the potential to be an electrode material for supercapacitors using hydrothermal carbonization [57, 58]. Despite having a higher rate of carbon synthesis, hydrothermal carbonization frequently results in materials with low porosity and SSA. Further research and development are needed for its commercial application. The schematic representation of the biochar preparation using hydrothermal carbonisation is presented in Fig. 2 (a). The Hydrothermal process was used to process biomass into valuable products. Usually, pyrolysis produces syngas, bio-oil, and solid biochar at elevated temperatures and atmospheric conditions. Alternatively, hydrothermal treatment involves processing biomass in a high-pressure, aqueous environment, producing primarily biochar along with some liquid byproducts. These conversion processes add value to biomass by transforming it into functional materials suitable for energy storage and various other applications. Notably, biochar stands out among the products due to its highly porous structure, making it especially advantageous for use as an electrode material in supercapacitor development.

### Torrefaction

Biochar, liquid oil, and syngas are the by-products of torrefaction, which are attained at a temperature range between 200 and 300 °C and atmospheric pressure [59]. The kind of wood used in the thermochemical processes, heating temperature and heating rate significantly influence the properties of the biochar. The hydrothermal method and slow pyrolysis are favoured in lieu of torrefaction, fast pyrolysis, and gasification processes for biochar production. However, slow pyrolysis is preferred instead of hydrothermal carbonization for alkaline biochar production. Slow pyrolysis introduces more aromatics into biochar than biochar produced by gasification or quick pyrolysis [60]. Most of the research focuses on slow pyrolysis and hydrothermal carbonisation techniques to increase surface areas and functional groups so that the final biochar can effectively remove potentially toxic elements (PTES).



(a)



(b)

**Fig. 2** (a) Production of syngas, bio-oil, and biochar via pyrolysis and hydrothermal techniques (b) Schematic representation of the biochar preparation techniques: pyrolysis, torrefaction, hydrothermal carbonization, and ionothermal carbonization and their experimental conditions

### Ionothermal carbonization

The one-stage ionothermal carbonisation process is used to create porous carbons from biomass. It is known that ionic liquids (ILs) are the safest solvents, and these have a low melting point (below 100 °C), are non-toxic and have excellent thermal stability, i.e., they are more resistant to changes

in their chemical or physical structure. The ILs are considered green solvents primarily due to their distinct characteristics, including chemical resistance, non-flammability, non-volatility, low vapour pressure and good thermal stability [61]. Due to these characteristics, ionic liquids have drawn much attention for use as both environmentally benign solvents and templates and porogenic agents in biochar formation [62]. To create biochar from carbohydrates,

Zhang et al. [63] adopted an ionothermal carbonization method with ionic liquids. The authors concluded that the generation of biochar material was higher, and it had noteworthy hierarchical holes, which were attributed to the porogenic effect produced by the iron (Fe) species discovered in the ILs. The schematic representation of the biochar preparation is presented in Fig. 2(b).

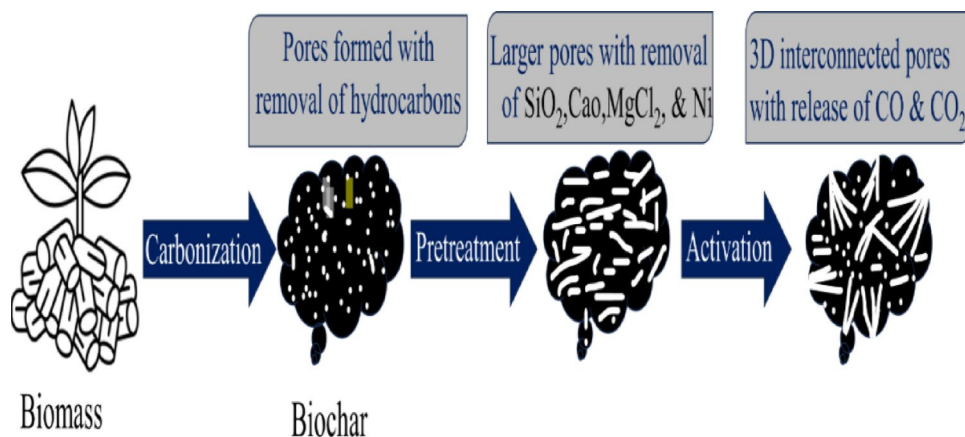
## Methods for activation of biomass-based porous carbon materials

The pyrolysis process, hydrothermal carbonisation method, and template approach are the only techniques to prepare porous carbon materials from biomass for supercapacitors. The produced carbon precursor must also be activated to acquire a large surface area and other physical and chemical properties. The various activation methods for biomass-based porous carbons are discussed below.

### Template methods

The core of the template method lies in the selection of various template materials. The hard and soft templates, along with other template techniques, are commonly used to synthesise biochar with specific pore size distributions. These template methods ensure high SSA, tunable pore size distribution, and exceptional electrical conductivity of the electrode materials [64]. When carbon materials are prepared using the template technique, the templates and biomass materials are carbonised. Later, residues are removed using an acidic or alkaline solvent to produce template-like porous carbon materials. Several biochar materials have outstanding electrochemical performance and have been created using the template approach for supercapacitors [65, 66]. Zeolite, mesoporous silica, and other commonly employed templates create carbon compounds with additional mesopore structures. The carbon materials using the template approach may have an adjustable pore structure.

**Fig. 3** Schematic illustrating the formation of interconnected porous structure of activated biochar derived from biomass through series of treatment techniques: carbonization, pretreatment, and activation



Nonetheless, the disadvantages of this approach are readily apparent, including the cost of the template materials and the length of the procedure.

### Carbon precursor activation

Two procedures, carbonization and activation, are most commonly used to create biochar using biomass as a precursor. First, a pore structure is generated during the carbonization process, which removes volatile contaminants (Fig. 3). Via the activator, the second step generates a large amount of pore structure and functional groups [67]. Following the activation process, the carbon precursor materials frequently display extensive SSA and well-established pore structure properties. It may be classified into physical and chemical activation, each with a different activation method, and these are discussed below in brief.

#### Physical activation

The physical activation occurs in the presence of gases like CO<sub>2</sub>, steam, air, and ozone [67]. The characteristics of the resulting biochar materials are affected mainly by the activation conditions, such as time and temperature, and the temperature is adjusted for the physical activation process at 600–1200 °C [67]. During activation, the following chemical reactions (1) and (2) take place:



During the process, carbon atoms will be eliminated due to the reaction, which facilitates the development of pore structure in the materials. Initially, the activator's active oxygen is consumed, removing leftover by-products and contaminants, further aiding in opening additional blocked pores. Ultimately, the carbon framework's surface area increases, in which CO<sub>2</sub> and H<sub>2</sub>O react as per the reaction

**Table 1** Influence of oxidising species on the idealised reaction mechanisms for Biochar production

Oxidizing agent	Idealized reaction mechanism	Endothermic/ Exothermic	Reference
Air/oxygen	$C + O_2 \rightarrow CO_2$	Exothermic	[70]
Steam/water	$C + CO \rightarrow CO + CO$ $C(O) \rightarrow CO$ $C + CO_2 \rightarrow 2CO$	Endothermic	[71]
Carbon dioxide	$C + H_2O \rightarrow CO + H_2$ $2C + H_2 \rightarrow 2C(H)$	Endothermic	[72]

**Table 2** Various Biochar activation methods and their characteristics

Biochar activation method and conditions			Acti- vating Agent	Elemental composition (wt%)					Pore Volume (cm <sup>3</sup> /g)	SSA (m <sup>2</sup> /g)	Reference
Feedstock	Carbonization	Activation		C	H	N	S	O			
Banana flesh	at 300 °C for 60 min	900 °C for 240 min	CO <sub>2</sub>	86.58	–	255	–	10.60	0.746	1415	[73]
Boabab fruit shells	at 160 °C for 960 min	800 °C for 120 min	H <sub>3</sub> PO <sub>4</sub>	84.51	–	1.41	–	14.01	0.47	911.70	[74]
K <sub>2</sub> CO <sub>3</sub> /biochar (2.5)	–	850 °C for 60 min	K <sub>2</sub> CO <sub>3</sub>	–	–	–	–	–	2.807	2312	[75]
Flaxseed residue (KOH/ biochar=3.5)	at 600 °C for 120 min	700 °C for 60 min	KOH	–	–	–	–	–	1.56	3326	[76]
Waste mangosteen peel (NaOH/biochar=3.5)	at 600 °C for 120 min	700 °C for 120 min	NaOH	–	–	–	–	–	–	2623	[77]
Aerobic granular sludge (50 M ZnCl <sub>2</sub> solution)	at 700 °C for 120 min	–	ZnCl <sub>2</sub>	–	–	–	–	–	0.086	852.41	[78]
Pine nut shell	at 500 °C for 15 min	900 °C for 75 min	Steam	85.93	2.15	0.57	0.32	8.47	0.62	956	[79]

scheme shown above (Eqs. (1) and (2)), resulting in the formation of new pore structures.

**Gas activation** Reactive gases serve as beneficial activators at high-temperature gasification, thereby enhancing the porosity and SSA of biochar materials [68]. At higher temperatures, biochar reacts with gas activation agents such as carbon dioxide, oxygen and water vapour [69]. This activation procedure efficiently removes impurities and leftovers of incomplete combustion from biochar. Table 1 also summarises the idealised reaction pathway for the activating agents CO<sub>2</sub>, O<sub>2</sub>, and water. Additionally, steam activation prevented biochar from using surface functional groups. The CO<sub>2</sub> efficiently enhances the surface area, i.e., SSA and pore volume of biochar.

### Chemical activation

The chemical activation technique activates the material in an inert environment using chemicals as an activating medium and occurs in the temperature range of 350, and 950 °C. This technique is widely employed due to its advantages, such as lower activation temperatures, quick activation time, high SSA, and controlled pore structure. However, using chemicals has certain drawbacks compared to physical activation, such as being expensive, difficult to recycle and reuse, and damaging equipment. Chemical reagents

often employed as activators include KOH, NaOH, K<sub>2</sub>CO<sub>3</sub>, salts such as ZnCl<sub>2</sub>, and acids like H<sub>3</sub>PO<sub>4</sub> [13, 67]. Table 2 lists some of the most often employed activators, including chemical and physical activation, as well as the properties of the biochar after preparation. Chemical activation can result in hierarchical pore structure and high SSA instead of physical activation, making it easier to create high-performance supercapacitor electrode materials. However, many expensive activators are needed for the chemical activation process, and most of them cause equipment corrosion. The product must be acid and water-washed during treatment to remove any remaining ash and inorganic material. The reaction method is inefficient and labour-intensive, significantly limiting its industrial use.

### Solid chemical activation

Biochar with a large surface area and clearly defined pore space are produced using activating chemicals. Potassium hydroxide (KOH), a key component, is necessary to enhance the surface area of charcoal. Though there are intensive research studies on experimental conditions and the biochar utilised in this procedure, the activation mechanisms are still unknown [80]. Water steam frequently forms from dehydrating biomasses when temperatures exceed 300 °C [81]. KOH must be heated to around 400 °C to decompose into K<sub>2</sub>O and water. Biomass is broken down to liberate carbon, which is then released through gas and water interactions to produce H<sub>2</sub> and CO<sub>2</sub> [82]. The environment's

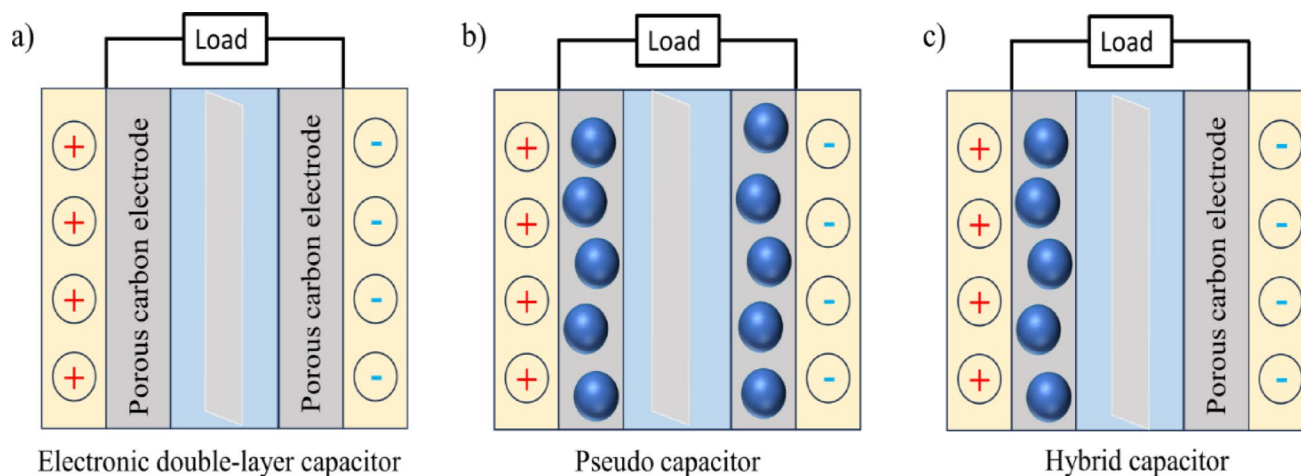
H<sub>2</sub>O and the CO released will react to create CO<sub>2</sub> and even more H<sub>2</sub> in the process. As the temperature increases above 800 °C and the ample H<sub>2</sub> and C reduce K<sub>2</sub>O (boiling point of 762 °C), interactions between CO<sub>2</sub> and K<sub>2</sub>O eventually result in the creation of K<sub>2</sub>CO<sub>3</sub>, which then provides metallic K [83]. The generated metallic K continuously catalyses the activation reaction, thereby constantly penetrating the pores and graphite layers [75]. Beyond those described in these equations, the different products have many additional interactions. The processes involving charcoal, KOH, and intermediates produce a high degree of porous network and, in turn, a high surface area. The ultimate pore network, comprising pore size, number, and surface area, is significantly impacted by experimental variables. These include activation agents, activation temperature, and mass ratios of different biochars and activation agents [84]. This involves directly adding KOH-treated biomass to the calcined biomass or impregnating it before the activation process starts. It is required to go through a low-temperature drying phase if biochar and KOH are produced through impregnation. The blend used for the activation process is heated in furnaces filled with inert gas, and the finished product is carefully washed with deionised water, followed by a mildly acidic solution before use to remove any soluble contaminants. The dried activated material exhibits a significant surface area and pore network [85]. Some drawbacks of activating biochar using activation chemicals include decreased end-product yields, production of greenhouse gases, corrosion of equipment, and increased industrial complexity. These factors must be taken into account if mass manufacturing of activated carbon is to be prevented for both environmental and financial reasons. Tea saponin was carbonised for 2 h at 550 °C, and chemical activation with KOH took place for 2 h at 750 °C [86]. They witnessed that the surface area would reach 1,550 m<sup>2</sup>/g [86]. Table 3 lists other chemical activators that are employed for biomass activation; these include HNO<sub>3</sub>, NaOH, ZnCl<sub>2</sub>, H<sub>2</sub>SO<sub>4</sub> in H<sub>3</sub>PO<sub>4</sub>, and K<sub>2</sub>CO<sub>3</sub> in the temperature range of 200 to 800 °C [74, 87–91].

## Supercapacitor as an electrochemical energy storage device

Supercapacitors are considered novel electrochemical devices as they have a high energy density, accept and deliver charge faster than conventional batteries, and can bear charge and discharge cycles [93]. Since high-energy batteries are frequently used for a variety of purposes, supercapacitors could have significant effects [89]. Supercapacitors may be utilised for shorter periods (a few seconds) and have higher output or uptake power densities (10 kW/kg) than regular batteries (5 Wh/kg) [94]. This, along with their quick discharge rates and potential for extremely high thermal stability, makes supercapacitors a viable technology with a wide range of applications [95]. Nonporous carbon electrodes serve as the foundation for most industrial supercapacitors because they are widely accessible, have a large surface area, are affordable, have good electrical conductivity, and offer a variety of surface functions. In the most recent study, this gadget performs better when carbon nanotube structures are used as electrodes [96]. The three primary supercapacitors are hybrid, pseudocapacitors, and electric double-layer capacitors (EDLC). Energy may be stored in a supercapacitor using pseudocapacitance (redox processes on the surface) and ion surface absorption (electrical double layer). Figure 4 shows two illustrations of this storage mechanism. The charge-discharge mechanism of the supercapacitor is the main reason for this. It permits a reversible ion accumulation on the electrode surface. Both the anode and the cathode are employed in their intended roles. The cathode (positive) and the anode (negative) can emit two kinds of ions. These ions spread through the electrolyte, forming a condensed layer parallel to the electrode surface [97]. In electrical double-layer capacitors (EDLCs), the charge is collected through electrostatic adsorption at the electrode/electrolyte interface [98]. Two layers of ions with different polarities emerge when the potential is applied across the contact boundary. These devices can achieve extremely high specific and volumetric capacitances due to their large surface area and thin double layer (0.5 to 1 nm).

**Table 3** Previously studied biomass for Biochar production with high BET surface area (m<sup>2</sup>/g) using various chemical activating agents and their activation conditions

Agricultural Waste	Chemical activating agent	Biochar activation conditions		BET Surface Area (m <sup>2</sup> /g)	Reference
		Carbonization	Activation		
Banana rachis	HNO <sub>3</sub>	at 600 °C for 120 min	at 200 °C for 120 min under 40 bar	859	[88]
Date palm kernels	H <sub>2</sub> SO <sub>4</sub>	Directly pyrolyzed at 600 °C for 90 min		741	[90]
Baobab shells	H <sub>3</sub> PO <sub>4</sub>	at 160 °C for 960 min	at 800 °C for 120 min	912	[74]
Waste bone	K <sub>2</sub> CO <sub>3</sub>	at 850 °C for 60 min	at 450 °C for 240 min	2312	[89]
Tea saponian	KOH	at 550 °C for 120 min	at 750 °C for 120 min	1550	[92]
Wheat husk	NaOH	at 400 °C for 240 min	800 °C until reaching the desired temperature	1200	[87]
Cassava petiole waste	ZnCl <sub>2</sub>	at 900 °C for 150 min	–	760	[91]



**Fig. 4** The energy storage mechanisms in the supercapacitors: (a) electronic-double-layer capacitor, (b) pseudo-capacitor and (c) hybrid capacitor

EDLCs do not use the Faradic techniques to store charge and do not transfer electrons from the electrolyte to the electrode, unlike batteries. In most cases, physical adsorption processes retain the charge, causing positive and negative ionic charges to build up on the electrode's surface and be equalised out by the electronic charge at the electrode's surface layer [99]. The electrode surface primarily controls the double-layer capacitance's electrical charge and is relative to the applied voltage. The amount of electrolyte and the electrode surface's structural makeup are two variables that may affect the formation of a double layer [53]. The aqueous or organic solvent-based electrolytes generally have an operating voltage per cell of less than 1–3 V. The electrode component of this EDLC is electrolyte-saturated and has a large surface area. A porous separator with the same electrolyte as the active substance sits between the active substance and the electrodes [100]. In pseudo-capacitors, the energy is generated through redox reactions in the electrolyte and electrode materials that create electrochemical capacitance in a Faradaic manner. The Faradaic process is finished by electro-sorption, reduction-oxidation reactions, and intercalation processes [90]. Pseudo-capacitors have larger capacitances and energy densities than EDLCs. On the contrary, these devices exhibit weak cycle stability and low power density [101]. Metal oxides and highly conducting polymers, such as polyaniline (PANI) and polypyrrole (PPy), are successfully used to improve the high pseudo-capacitive behaviour of electrode supercapacitors [102]. Due to little surface area dependence, their specific capacitance is higher than the EDLC mechanism [103]. The maximum electrochemical capacitance of an electrode is a crucial parameter for an electrochemical energy system. It was determined that the electrochemical energy system's specific capacity, energy density, power density, and electrochemical capacitance should be investigated. Further, surface reactions on electrodes have the drawback of slowly deteriorating the

material, raising the possibility of failure, and lowering cycle stability. A small portion of an electrostatic double-layer and pseudo-capacitive mechanisms are used to store charge. The capacitance of a pseudocapacitor system is much greater than that of a straightforward double-layer technique, owing to the possibility that the reactant molecules in the pseudo-capacitor bulk layer would donate one or more charges to the electrode surface. Although pseudocapacitors have a larger capacitance than EDLCs, they can have substantially lower charge-discharge lifetime stability [104]. Redox processes can cause the deterioration of polymeric materials, resulting in energy loss over time. Scientists are combining pseudo-capacitive and electrochemical capacitive substances to produce electrodes that have greater benefits than the separate materials as a result of the recent focus on hybrid capacitors. The Faradaic and double-layer materials have been used to create composite capacitor electrodes that have shown enhanced capacitance and stability (Fig. 4). Generally, three types of supercapacitors: (a) electric double-layer capacitors (EDLCs) store energy via electrostatic charge separation on porous carbon electrodes, (b) pseudocapacitors utilise fast redox reactions for charge storage, and (c) hybrid capacitors combine both mechanisms, using a pseudocapacitive and a carbon electrode. Each type varies in charge storage mechanism, energy density, and performance. The hybrid capacitors made of lithium-ion are electrostatic and electrochemical capacitance capacitors [94]. Higher specific energy density balances off the loss of high specific power in a hybrid capacitor. These materials exhibit poor conductivity, high cost, and toxicity, thereby limiting their applications despite their high pseudocapacitance [105]. With low cost, high specific capacitance, good conductivity, and ease of manufacturing, conductive polymers are more commonly used as hybrid capacitors [106].

Machine learning (ML) techniques have been employed to investigate the impact of surface chemistry on the

electrochemical characteristics of carbon materials derived from biomass. These methods enable the prediction and enhancement of the performance of energy storage devices. By utilising ML models, it becomes feasible to anticipate the capacitance of graphene-based supercapacitors doped with heteroatoms, thereby establishing the influence of heteroatom-doping [107]. Notably, the XGBoost model has demonstrated superior predictive capabilities, as evidenced by its low mean root mean square error and coefficient of determination close to 1 [108]. This particular model has successfully been applied to forecast the cyclic voltammetry behavior of supercapacitors composed of cobalt-doped ceria/reduced graphene oxide (Co-CeO<sub>2</sub>/rGO) fractal nanocomposites [109]. The development of machine learning models, such as k-nearest neighbors' regression (KNN), decision tree regression (DTR), Bayesian ridge regression (BRR), and artificial neural network (ANN), has relied on experimental data extracted from over two hundred research papers. These models have been utilised to predict the specific capacitance of electrode structures based on graphene [110]. Additionally, ML techniques have been employed to forecast the capacitance and remaining useful life (RUL) of supercapacitors, offering a promising avenue for performance prediction and addressing system failure [111]. Furthermore, ML algorithms have been employed to establish correlations between intrinsic features of supercapacitors, such as electrode materials and electrolyte types, and cyclic stability. These correlations offer valuable insights for the development of novel material combinations. The ANN models have exhibited a high correlation coefficient value of 0.99, indicating a robust predictive capability for the electrochemical properties of electrode materials [112]. Furthermore, ANN modelling has been evaluated to elucidate the influence of the activation procedure, structural features, electrode synthesising procedure, and operational conditions on the capacitive performance of carbon-based electrodes [113]. Activated carbon derived from biomass waste has been employed as an active material in textile-based electric double-layer capacitors, achieving a specific capacitance of 80 mF/g, energy density of 5.36 mWh/Kg, and power density of 4.87 W/Kg [114]. These findings offer valuable insights for the development and optimisation of supercapacitors based on biomass.

## Role of carbon in supercapacitors

Carbon-based electrode materials are widely used because of their low cost and widespread accessibility. Additionally, altering the pore structure, functionality, and surface area of carbon compounds used in energy storage devices is simple. The electrostatic attraction of electrolyte ions to the surface

of the electrode's substance is used in supercapacitors to store charge; thus, the electrode material must have the best pores that match the size of the electrolyte ions [115]. Surface functionalities such as hydroxyl, carboxylic, nitrogen, and phosphorus groups contribute to pseudo-capacitance through electrochemical redox reactions, enhancing charge storage capacity. The degree of graphitization is an essential criterion for carbon-based materials utilised in supercapacitors since it boosts electrical conductivity [116]. The carbon framework's structural integrity ensures long-term cycle stability and capacitance retention.

Carbon materials utilised in supercapacitors encompass biomass-derived carbon composites (BDCC), carbon-based substances, porous carbons derived from various sources like biomass, polymers, lignite, metal salts, and melamine, as well as carbon nano-onions (CNOs) [117]. BDCC, carbon-based materials, and porous carbons have exhibited notable characteristics, including high specific capacitance, long-term cyclic stability, and high surface area, rendering them suitable for implementation in supercapacitors [118]. When functionalised and modified with resins, CNOs have demonstrated the ability to significantly amplify the overall pore volume and enhance the electrochemical properties of the materials. The carbon material derived from CNO, resorcinol, and melamine (RFM-CNO-C) showcased the highest specific capacitance and sustained performance after cycling. The effectiveness of supercapacitors is heavily influenced by the carbon material employed as the electrode [119]. Carbon-based electrodes are immensely desirable due to their cost-effectiveness, abundance, and capacity to manipulate conductivity and surface area. The surface attributes of carbonaceous materials, such as their porous structure and hydrophilicity, exert a profound impact on the electrochemical performance of supercapacitors. Active carbons represent the most commonly utilised carbonaceous electrode materials due to their commendable chemical stability, conductivity, and accessibility. Nonetheless, their energy densities still fall short of practical demands. Surface modification and doping of carbonaceous materials allow for the optimization of their pore size, structure, conductivity, and surface attributes while also introducing supplementary pseudocapacitance, thus enhancing their performance. Furthermore, the amalgamation of reversible redox mediator addition and heteroatom-doping of carbon-based electrode materials has been shown to heighten the electrochemical performance of supercapacitors. Novel high-capacity electrode materials and electrolytes, including ionic liquids and gel polymers, have also been devised to ameliorate the energy densities of supercapacitors.

## Application of activated biochar in supercapacitors

Diverse techniques have been utilised to make biochar with desirable characteristics such as high SSA, high mesopore content, hierarchical pore structure, and high energy density under capacity efficiency and rate stability criteria.

### High specific surface area biochar materials

KOH has been established to be an efficient activator for producing biochar among the many available activating chemicals. Maintaining a mass ratio of KOH/biochar of 4:1 or 3:1 has resulted in the highest SSA. A biochar material made from lotus leaves was reported by Lu et al. [120] to have outstanding electrochemical performance. The lotus leaves were initially pyrolysed at 700 °C to extract the carbon in a tubular furnace. The harvested biochar was then heated to 700 °C and activated with KOH (1:3 wt. ratio). The resulting biochar has a high SSA of 2350.8 m<sup>2</sup>/g and a 1.37 cm<sup>3</sup>/g pore volume. It demonstrated a high specific capacitance of 478 F/g as a supercapacitor electrode at a current density of 1 A/g. The built symmetric supercapacitor device also showed an extended life cycle, with 89.10% capacitance retention after 5000 cycles at 5 A/g in a two-electrode system. Biochar electrode material was prepared by Li et al. [76] using KOH generated from flaxseed residue. Under inert gas, the precursor was carbonised at temperatures between 600 and 800 °C. Further, the products were activated at 700 °C in an argon environment by mixing them 1:4 with KOH. The biochar material from flaxseed residue had a high SSA of 3230 m<sup>2</sup>/g; micropore volume comprised 70.10% of the overall volume. The biochar substance showed specific capacitance in KOH and H<sub>2</sub>SO<sub>4</sub> electrolytes of 369 and 398 F/g, respectively, and 98.10% capacitance retention after 10,000 cycles. Shang et al. [121] described the biochar substance made for supercapacitors using carbonization and activation procedures. KOH developed biochar substances by employing walnut shells as a feedstock. The precursor was carbonised at 500 °C in an inert tube furnace. The biochar material made from walnut shells has a high specific surface area (SSA) of 3577 m<sup>2</sup>/g and a total pore volume of 2.19 cm<sup>3</sup>/g. Additionally, it showed a specific capacitance of 330 F/g at 0.1 A/g, had a good pore size distribution, had a sheet-like structure, and had an excellent capacitance retention of 81% in a two-electrode system with 6 M KOH. The activators, K<sub>2</sub>CO<sub>3</sub> and KHCO<sub>3</sub>, biochar substances showed specific capacitance of 369 and 398 F/g in KOH and H<sub>2</sub>SO<sub>4</sub> electrolytes, respectively and 98.10% of the capacitance retention after 10,000 cycles as a result of an ongoing study into these materials. Yuan et al. [122] studied chitosan-derived hybrid carbon

composites. KHCO<sub>3</sub> gave the gelatin-modified chitosan-derived biochar a distinctive tangerine pith-like shape with an SSA of 927.17 m<sup>2</sup>/g. The biochar electrode material with a distinct pith-like morphology had a high specific capacitance of 331 F/g in a 6 M KOH electrolyte at 1 A/g and 90% capacitance retention after 10,000 cycles at 10 A/g.

The biochar material made from chitosan demonstrated an energy density of 34 Wh/kg, significantly exceeding most commercially available devices with a power density of 900 Wh/kg. The power density is influenced by the entire pore volume and SSA, which is connected to ion storage capacity. However, the development of biochar materials is constrained by the low volumetric density of electrode materials. A P-doped porous carbon material based on *Elaeocarpus Tectorius* was suggested by Nirosha et al. [123] to generate porous P-doped carbon materials for use as high-performance electrode materials in supercapacitors; the precursor was activated using a straightforward one-step procedure using the activating agent H<sub>3</sub>PO<sub>4</sub>. In a 1 M H<sub>2</sub>SO<sub>4</sub> electrolyte, the carbon reaction that was achieved at 900 °C had a high gravimetric capacitance of 385 F/g at 0.2 A/g and volumetric capacitance of 543 F/cm<sup>3</sup> at 0.2 A/g. The device revealed a fantastic potential for supercapacitors with a specific energy of 33.8 Wh/kg and a specific power of 648 W/kg. Cotton-based biochar materials were used as supercapacitor electrodes [124]. The discarded cotton was first carbonised at 600 °C in an inert environment, and then the resulting carbon was activated with KOH at 950 °C in an N<sub>2</sub> atmosphere. The synthetic biochar had an SSA of 1893 m<sup>2</sup>/g and a volumetric capacitance of 87 F/cm<sup>3</sup> at 1 A/g. It also gives a greater volumetric energy density of 30.94 Wh/L in a voltage window of 3.2 V, achieving high active mass loading and the widest voltage window simultaneously. Furthermore, the biochar materials made from cotton showed high endurance of up to 15,000 charge-discharge cycles at 4 A/g. The concept of electrochemical double-layer electrodes states that a high SSA offers added ions in the electrolyte solution adsorption sites, which increases the specific capacitance of the electrode material. The SSA and the specific capacitance are not linear because most SSA in biomass-based porous carbon materials is given by tiny diameter holes, which electrolyte ions cannot fully use with larger diameters. An electrode material with an excellent specific capacitance may not have an electrode material with a bigger SSA, but a higher SSA may bring about an optimum specific capacitance. A high specific surface area (SSA) is sufficient but not necessarily essential for achieving high specific capacitance. The electrochemical stability of a few electrode materials with considerable SSA is listed in Table 4. Creating high-performance electrode materials depends on optimising the use of SSA.

## Mesopore biochar materials

Mesopores with a wide pore diameter as a conduit for the fast transfer of electrolyte ions must be added to carbon materials to increase the biochar's SSA utilisation rate. Zhang et al. [137] reported the preparation of electrodes with high specific capacitance and high energy density using Chinese date biomass. The precursor was pyrolyzed at 700 °C with N<sub>2</sub> to produce the carbonised product. The product was then activated in various temperature ranges of 500–700 °C under N<sub>2</sub> environment using a KOH activator in various ratios. The obtained Chinese date-based carbon had an SSA of 1941 m<sup>2</sup>/g, pore volume of 0.85 cm<sup>3</sup>/g, and mesopore volume of 0.26 cm<sup>3</sup>/g. It was activated at 600 °C with a mass ratio of KOH/biochar of 1:4. Under various reaction conditions, mesopores had a substantially larger volume ratio than other biochar substances. It displayed a greater specific capacitance of 518 F/g at 0.5 A/g of current density compared to the other and a rate performance of 40 F/g at 70 A/g. Zhang et al. [138] created honeycomb-like carbon nano-sheets with controllable mesopores using mildly modified coal tar pitch as a precursor. The solvent extraction, light modification, and activation procedure were used to prepare the carbon materials. The extracted precursor and KOH are combined in a 1:3 ratio and activated at 700 °C in an N<sub>2</sub> environment. The materials had a specific capacitance of 411.20 F/g and high conductivity due to the honeycomb nanosheet characteristics and quantity of mesopores with a 2–5 nm diameter. It had a high energy density of 12.30 Wh/kg at a power density of 249.50 W/kg as an integrated symmetrical flexible all-solid-state supercapacitor with PVA/KCl membrane electrolyte. Jiang et al. [139] created cotton-based porous carbon while maintaining the original hollow-tubular fibre structure through carbonization and KOH-KNO<sub>3</sub> activation. It was possible to obtain carbon material with a large surface area (1508 m<sup>2</sup>/g) and a clear mesoporous structure with a diameter of 9.18 nm. This type of carbon material displayed

a high specific capacitance of 278 F/g at 1 A/g and 208 F/g at 100 A/g as well as an exceptionally long cycle life, which may be related to its unique hollow tubular morphology and activated porous structure. Through carbonization and activation processes, Zhu et al. [140] suggested using egg white to synthesise the nitrogen-doping biochar materials. The egg white-based biochar materials comprised a three-dimensional honeycomb structure and an SSA of 2918 m<sup>2</sup>/g. The network of micro- and mesopores might improve the ion diffusion channel in the electrolyte and expand the region in contact with the electrode. Additionally, it demonstrated 91.70% capacitance retention after 10,000 cycles in a three-electrode setup and a specific capacitance of 335 F/g at 0.5 A/g. Yang et al. [141] created hierarchical biochar materials based on wood sawdust through hydrothermal carbonization and activation methods. The structure of the macropores and mesopores was then created during activation with the help of KOH. The biochar materials provided a high energy density of 17.75 Wh/kg at a power density of 436 W/kg, great cycle stability, and a specific capacitance of up to 303 F/g at 1 A/g. Activators like ZnCl<sub>2</sub> and NaOH greatly aid mesopore development in charcoal materials. Among the different chemical agents that activate, ZnCl<sub>2</sub> has well-known capabilities as a dehydrating agent and may be utilised to create better porous structures [142]. Zhang et al. [143] proposed wheat bran-based biochar materials using NaOH activation. The produced biochar materials with hierarchical micropores and mesopores showed an SSA of 2562 m<sup>2</sup>/g when the mass ratio of the precursor and NaOH was 1:4. It provided cycle stability of 94% for 50,000 cycles and a specific capacitance of 294.3 F/g at 0.5 A/g as the supercapacitor electrode. Ahmed et al. [144] described using a one-step thermal activation procedure to create high-specific area biochar materials from the pollen of *Butnea monsperma* flowers. First, a concentrated solution of ZnCl<sub>2</sub> in varied strengths was added to the feedstock. The mixture was then dried in a vacuum oven at 100 °C

**Table 4** Electrochemical performance of electrodes made using high-specific surface area Biochar materials. Here, BHMIMBF<sub>4</sub>/AN is 1-butyl-3-methylimidazolium tetrafluoroborate/acetonitrile electrolyte

Raw material used for biochar production	Chemical activating agent used	BET surface area (m <sup>2</sup> /g)	Electrolyte	Reported current density (A/g)	Capacitance (F/g)	Reference
Cotton seed husk	KOH	1694	6 M KOH	0.5	238	[125]
Poplar anthers	KOH	3639	6 M KOH	0.5	362	[126]
Sisal	KOH	2289	6 M KOH	0.5	415	[127]
Rice husk	KOH	3263	6 M KOH	0.5	315	[128]
Rice husk and Crab shells	KOH	3557	6 M KOH	0.5	474	[129]
Castor shell	KOH	1527	6 M KOH	1	365	[130]
Cotton stalk	KOH	1964	1 M Na <sub>2</sub> SO <sub>4</sub>	0.2	254	[131]
Anthracite	KOH	2814	1 M Na <sub>2</sub> SO <sub>4</sub>	0.5	325	[132]
Shaddock endothelium	KOH	1265	1 M BHMIMBF <sub>4</sub> /AN	0.2	550	[133]
Cellulose	NaOH	1588	6 M KOH	0.5	288	[134]
Rose flower	KOH/KNO <sub>3</sub>	1980	6 M KOH	1	350	[135]
Cornstalk	K <sub>2</sub> C <sub>2</sub> O <sub>4</sub> .H <sub>2</sub> O	2054	1 M Na <sub>2</sub> SO <sub>4</sub>	0.5	461	[136]

for 24 h. The dried mixture was then pyrolyzed in a tube furnace with an environment of  $N_2$  at 800 °C. The carbon material produced significant results for a total pore volume of 0.77  $cm^3/g$  and SSA of 1422  $m^2/g$ . After 10,000 cycles in an aqueous electrolyte, the electrochemical properties of biochar made from *butnea monsperma* flower pollens were similar to 130 F/g and had better capacitance retention of 99%. Sun et al. [145] developed biochar substances with numerous nitrogen-doped mesopores made from left-over maize silks. The SSA and pore volume of the biochar were 1764.80  $m^2/g$  and 2.0  $cm^3/g$ , respectively. After 5,000 cycles, the biochar electrode still had 99.20% of its initial specific capacitance and delivered a specific capacitance of 358 F/g at 0.5 A/g. The biochar electrode also produced a high energy density of 17.8 Wh/kg and a specific capacitance of 260.80 F/g, utilising  $H_2SO_4$  solution with alizarin red and bromoamine acid as an advanced electrolyte. Mesoporous structures can enhance the utilization rate of specific surface area (SSA) in porous carbon materials, often formed through pore expansion. However, this process can reduce overall SSA and limit the availability of active sites for energy storage, ultimately compromising the material's electrochemical stability.

### Hierarchical biochar material

The main source of SSA in the majority of biochar products is micropores. The micropore structure greatly decreases the internal active sites and the rate at which electrolyte ions diffuse through the electrode material. Typically, the growth of micropores results in the formation of mesopore structures. The SSA and the active sites of the porous carbon material would diminish once the micropore structure was sacrificed. To generate biochar electrodes with high specific capacity and electrical conductivity, hierarchically porous carbon materials with connected pore structures must be fabricated, and the ratio of macropores, mesopores, and micropores must be logically designed. Among the most intriguing supercapacitor electrodes are carbons generated from biomass with hierarchical porous architectures. Wu et al. [146] suggested using KOH activation to create hierarchical *albizia* flower-based biochar micro-rods. The generated biochar material had a special porous microrod structure and self-doped nitrogen. It was combined with KOH in a mass ratio of 1:3 and activated at 900 °C. A total pore volume of 1.47  $cm^3/g$  and a high SSA of 2757.63  $m^2/g$  were witnessed in the biochar. Due to these benefits, it proved excellent cycle stability with 97% capacitance retention after 5000 cycles and had a high specific capacitance of 390 F/g at 1 A/g. Furthermore, it provided a specific energy density of 26.30 Wh/kg at a power density of 429 W/kg in a 1 M  $Na_2SO_4$  electrolyte [146]. Zhou et al. [147] created a

biochar material resembling a honeycomb using bristlegrass seeds. The 3D interconnected hierarchical pore structure, high SSA, and multi-heteroatom doping of the produced biochar as an electrode demonstrated a high specific capacitance of 391 F/g at 0.5 A/g. Additionally, it offered a 97.20% capacitance retention over 10,000 cycles, along with good rate capability and cycling stability. The supercapacitor electrode demonstrated a high energy density of 20.15 Wh/kg at a power density of 500 W/kg in 1 M  $Na_2SO_4$  aqueous electrolyte. To synthesise biochar with a hierarchical porous structure for an ultrahigh-rate supercapacitor, Guo et al. [148] utilised a dual-activation approach. By using  $KMnO_4$  and KOH to activate cotton stalks simultaneously, interconnected porous carbon was produced. This carbon material in a two-electrode system has demonstrated superior cycle stability with an electrolyte comprising 6 M KOH and a high capacitance of 318 F/g at 1 A/g. The symmetric two-electrode supercapacitor also demonstrated a maximum energy density of 19.90 Wh/kg at a power density of 379 W/kg using 1 M  $Na_2SO_4$  as the electrolyte [148]. Cai et al. [149] created hierarchical porous carbon nanosheets from *Moringa oleifera* stems using a one-step pyrolysis method. The as-prepared biochar material has a special porous nanosheet shape, a high specific surface area (SSA) of 2250  $m^2/g$ , a big pore volume of 2.3  $cm^3/g$ , the right amount of porosity, and heteroatom doping of nitrogen and oxygen. It displayed a high specific capacitance of 283 F/g at 0.5 A/g and a capacitance retention of 72% at a very high current density of 50 A/g. Furthermore, the three-electrode system has demonstrated exceptional long-term cycling stability. Khan et al. [135] described using naturally withering rose blossoms to create biochar using a KOH/ $KNO_3$  activation process with hierarchically arranged pores. The resulting carbon material demonstrated excellent conductivity, a high SSA of 1980  $m^2/g$ , and a hierarchical pore structure. It demonstrated exceptional electrochemical stability in a three-electrode system as the electrode, with a specific capacitance of 350 F/g at a current density of 1 A/g, an exceptional rate ability of 165 F/g at 150 A/g, and a capacitance retention of 95.40% after 140,000 cycles at 100 A/g. With the crosslinking effect of cellulose, hemicellulose, and lignin, biomass precursors' effectiveness in tailoring the pore structure has become challenging. Hydrothermal treatment of biomass is an efficient method that assures disruption of the components' crosslinking, resulting in an effective pore structure [19]. Chen et al. [150] recommended using tremella as the precursor to manufacture biochar material with a wire-like shape using KOH activation. First, the tremella was placed in an airtight autoclave for 12 h at 120 °C using the hydrothermal carbonization procedure. The carbon products were washed several times and immersed in various aqueous KOH concentrations. After drying, the KOH and

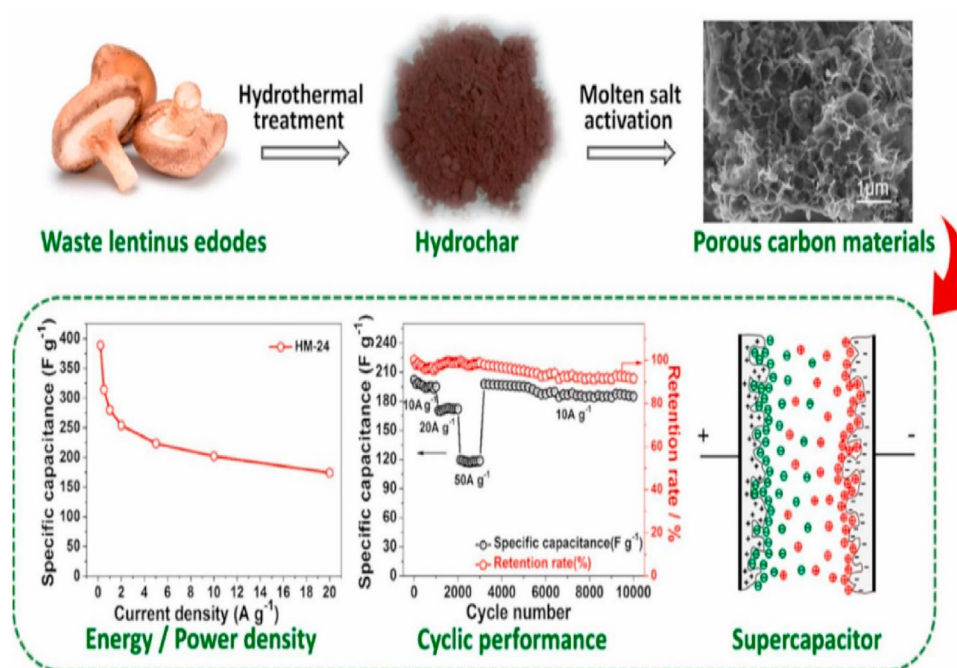
tremella mixtures were activated at 700 °C in N<sub>2</sub>. The as-obtained biochar electrode showed good cycle stability, a high capacitance retention of 250.30 F/g at a current density of 20 A/g, and a specific capacitance of 299.30 F/g at 0.5 A/g in aqueous electrolyte. Tang et al. [151] created waste-based hierarchical biochar materials using the *lentinus edodes* plant. Figure 5 depicts the synthesis procedure for porous carbon electrodes using waste *lentinus edodes* and its primary electrochemical activity. The hydrothermal carbonization process was used to carbonise the hierarchical biochar materials, and they were then activated in molten Na<sub>2</sub>CO<sub>3</sub>-K<sub>2</sub>CO<sub>3</sub>. Numerous tiny pore structures were created on the surface of the biochar after hydrothermal treatment. These pore structures were formed at the end of the hydrothermal carbonization. The continual activation of the molten salt approach to create the hierarchical pore structure was greatly aided by the presence of this type of pore structure. The biochar was obtained as-is and had a hierarchical pore structure and a high SSA of 1144 m<sup>2</sup>/g. Additionally, the biochar electrode had a specific capacitance of 389 F/g at 0.2 A/g and an exceptional rate capacity, with capacitance remaining 174 F/g at 20 A/g in a three-electrode system. These characteristics were due to oxygen- and nitrogen-containing functional groups that were produced at the biochar surface. The built symmetric supercapacitor also provided a high specific capacitance of 329 F/g at 0.2 A/g, a high energy density of 45.69 Wh/kg, and outstanding cycling stability after 10,000 cycles, maintaining 90.30% of the starting capacitance at 5 A/g. Through hydrothermal and oxidative pre-treatment, a more controlled hierarchical porous carbon may be produced, depending on the unique

structure of the biomass itself. The hierarchical porous carbon outperformed ultra-high SSA and mesoporous carbon materials in terms of electrochemical performance owing to a balanced ratio of macro, meso, and micropores.

### Surface modification of Biochar materials

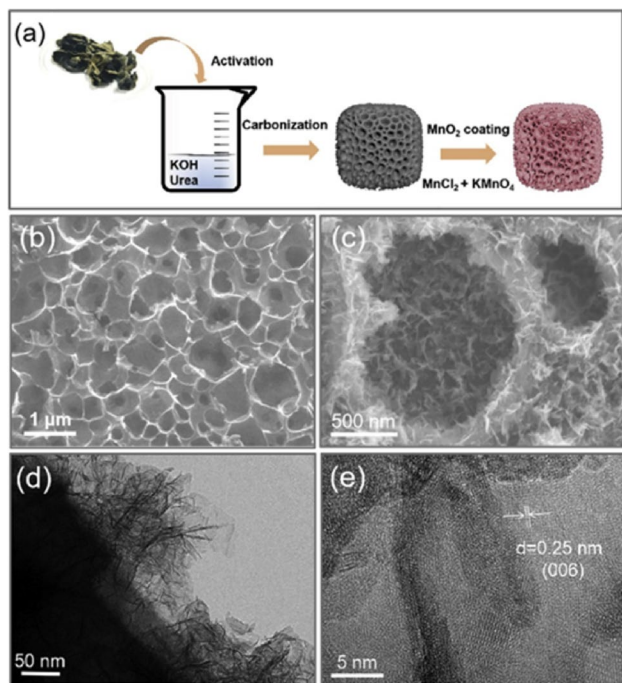
Several research studies have demonstrated that introducing heteroatoms to carbon-based materials' surfaces can increase supercapacitors' capacitance even further. For instance, adding N, O, and P atoms to the surface can enhance the wettability between the electrolyte solution and the electrode material, thereby facilitating the effective interaction between the electrode and the ions of the electrolyte solution. O and N atoms as functional groups on porous carbon material exhibit acidic or alkaline properties, improving the interaction between the electrode and the electrolyte. Wan et al. [153] proposed an N, O, S multi-heteroatom-doped hierarchical biochar made from chestnut shells using melamine as the activating agent. The synthetic biochar materials had an SSA of 691.80 m<sup>2</sup>/g and included heteroatoms with concentrations of 3.79 at% N, similar to 13.35 at% O, and similar to 0.52 at% S. They had an appropriate degree of graphitization and linked micro- and mesopores. In a three-electrode setup, the produced biochar displayed a high specific capacitance of 402.80 F/g at a current density of 0.5 A/g and an outstanding rate capability of 45.30% capacitance retention at 100 A/g in KOH electrolyte. It also demonstrated exceptional cycling stability with a 96.60% capacitance retention after 20,000 cycles. Wang et al. [154]

**Fig. 5** Schematic illustration of the synthesis process of hierarchical porous carbon materials using waste *lentinus edodes* and their electrochemical performance for a supercapacitor (adopted from Li et al. [152])



proposed a biochar material made from durian shells that is N, O, and P multi-heteroatom-doped. The biochar materials had numerous heteroatoms doped, a moderate SSA, and a pore shape predominantly made up of micropores. The supercapacitor's specific capacitance is 184 F/g at 0.5 A/g current density. It displayed remarkable cycle stability with a retention rate of 88% after 10,000 cycles. Additionally, in  $\text{H}_2\text{SO}_4$  and KI aqueous solution, multi-heteroatom-doped biochar had a capacitance of 560 F/g at a current density of 2 A/g. The O, N, and S-doped hierarchical biochar material for supercapacitors was synthesised without the need for an activating agent using kraft lignin as the precursor [155]. The technique of preparation was effortless, affordable, and sustainable. The biochar made from kraft lignin had an SSA of  $1307 \text{ m}^2/\text{g}$ , hierarchical porous structures, and a significant proportion of multi-heteroatoms co-doping (9.84–19.91 wt%). The biochar electrode was successfully synthesised and provided a high specific capacitance of 244.50 F/g at 0.2 A/g, satisfactory conductivity, 81.80% capacitance retention of initial capacitance at 40 A/g, and outstanding cycling stability of 91.60% retention over 10,000 cycles. The supercapacitor demonstrated an energy density of 66.80 Wh/kg in the aqueous electrolyte at a power density of 1.75 kW/kg, and it was still maintained at 32.20 Wh/kg with an ultrahigh power density of 40 kW/kg. Through a straightforward one-step eco-friendly process, Yan-Xia et al. [156] suggested an

N-doped cotton-based porous carbon material. The synthesized biochar exhibited a specific surface area (SSA) of  $480 \text{ m}^2/\text{g}$  and achieved a maximum specific capacitance of 252 F/g at a current density of 1 A/g in a 1 M  $\text{H}_2\text{SO}_4$  electrolyte. Additionally, it retained 94% of its capacitance after 10,000 charge-discharge cycles at 15 A/g, demonstrating excellent long-term electrochemical stability [156]. Pseudocapacitors may be added to electrochemical double-layer capacitors to increase the biomass-grade biochar materials' capacity further. Adding heteroatoms and metal ions is the foremost technique. Among a variety of functional materials,  $\text{MnO}_2$  is employed due to its benefits of sustainability, affordability, and environmental friendliness. Li et al. [152] proposed an agaric-based N-doped porous carbon composite material with stacked  $\text{MnO}_2$  nano-sheets. Figure 6 (a and b) depicts the  $\text{MnO}_2$  deposited on N-doped activated porous carbon ( $\text{MnO}_2@N\text{-APC}$ ) synthesis procedure, scanning electron microscopy (SEM), and transmission electron microscopy (TEM) images. Figure 6 (a) illustrates the swelling of agaric in the presence of KOH and urea, as well as the subsequent carbonisation and loading of  $\text{MnO}_2$ . Figure 6 (b) depicts a well-defined 3D porous structure with interconnected pores that serve as a suitable site to load  $\text{MnO}_2$ . Also, pores serve as a track for rapid electron transfer. Figure 6 (c–e) portrays the SEM and TEM of  $\text{MnO}_2$ -loaded N-APC and HRTEM revealing the crystalline nature of the composite [76].

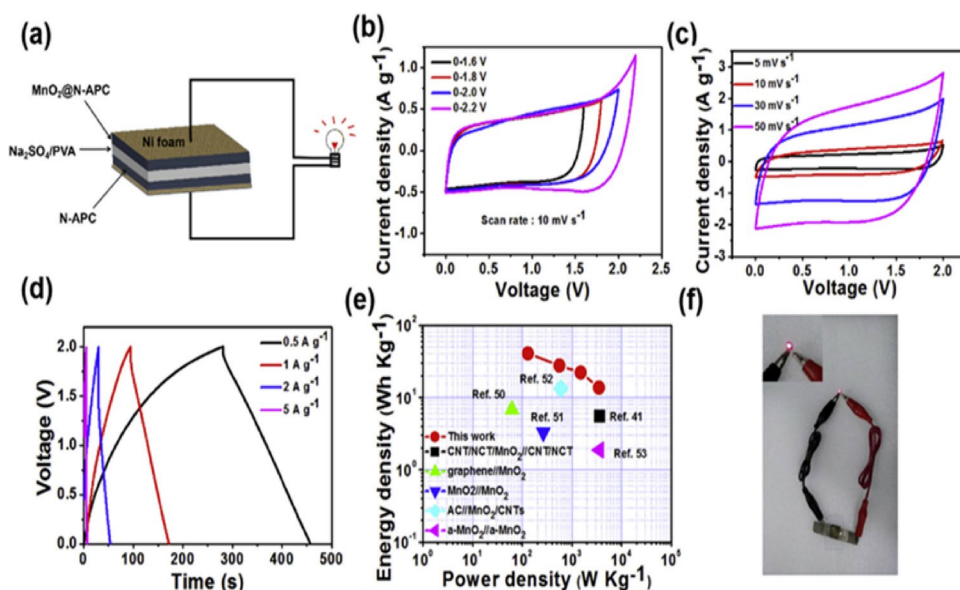


**Fig. 6** (a) Schematic of the synthesis procedure for  $\text{MnO}_2@N\text{-APC}$  composite. (b) SEM images of the N-APC sample depicting an interconnected 3D structure. (c) SEM image of  $\text{MnO}_2$  loaded on N-APC. (d) TEM image of the  $\text{MnO}_2@N\text{-APC}$  composite. (e) Crystalline nature of  $\text{MnO}_2$  nanosheets (adopted from Li et al. [76])

Further, an asymmetric supercapacitor was fabricated with  $\text{MnO}_2@N\text{-APC}$  and N-APC as positive and negative electrodes in 1 M  $\text{Na}_2\text{SO}_4$  electrolyte, shown in Fig. 7(a). This supercapacitor has a large voltage window that can attain a value of 2.0 V, suggesting a higher energy density revealed from Fig. 7(b). At the same time, Fig. 7(c) reveals that the composite has ideal capacitance characteristics. Figure 4(d) reveals the symmetrical nature of the charge discharge curves, corroborating the Cyclic voltammetry (CV) tests. The Ragone plot ( Fig. 7(e) ) reveals the device's better performance. Figure 7(f). demonstrates that this asymmetric supercapacitor device can glow a red light-emitting diode (LED) at 2 V.

The resulting biochar material, which had an extremely high SSA of up to  $2,250 \text{ m}^2/\text{g}$ , provided a sizable contact area for inserting  $\text{MnO}_2$  nano-sheets. The biochar material's distinctive three-dimensional pore structure morphology promoted the transmission and diffusion of ions in the electrolyte solution. With a specific capacitance of 330 F/g at 1 A/g, the  $\text{MnO}_2@N\text{-APC}$  composite hence demonstrated exceptional electrochemical performance. The supercapacitor device showed remarkable performance with a big voltage window of 2 V, an energy density of 28 Wh/kg

**Fig. 7** (a) Schematic of the asymmetric supercapacitor structure. (b) CV curves of the asymmetric supercapacitor with a scan rate of 10 mV/s under different voltage windows. (c) CV curves, (d) GCD curves and (e) Ragone plots of the asymmetric supercapacitors, (f) glow of LED light by the asymmetric supercapacitor. (adapted from Li et al. [76].



at a power density of 560 W/kg, and a negative electrode made of biochar material. Bagasse was employed by Zhou et al. [157] to create an ultrathin  $\text{MnO}_2$  nanosheet/activated porous carbon composite ( $\text{MnO}_2/\text{AC}$ ). KOH activation was used to activate the bagasse-based carbon at 800 °C in an Ar environment. Hydrothermal carbonisation was used to create the  $\text{MnO}_2/\text{AC}$ . The  $\text{MnO}_2/\text{AC}$  composite electrode demonstrated a specific capacitance of 492.50 F/g at 1 A/g and 78.40% capacitance retention after 5,000 cycles using AC material as the negative electrode. Compared to  $\text{MnO}_2/\text{AC}$ , the AC electrode had a higher specific capacitance of 89 F/g at 1 A/g and cycling stability of 89% capacitance retention after 5000 cycles. Polymer compounds may also be doped onto materials made of porous carbon. A highly promising polymer chemical for enhancing pseudo-capacitance, according to studies, is polyaniline. Due to their widespread study, facile synthesis, and strong chemical and environmental stability, these compounds are readily available. Using cross-linked carbon nanosheets and polyaniline, Li et al. [158] created a composite in three dimensions. High surface area and an interconnected pore structure in biomass-based carbon materials with graphene-like properties might serve as embedding sites for PANI to induce pseudo-capacitance. This mixture could facilitate the transit and diffusion of electrolyte ions inside the electrode material and enhance specific capacitance and conductivity. The PANI/CCNs composite electrode performed well with a high specific capacitance of 444 F/g at 0.1 A/g and 340 F/g at 5 A/g current densities. A specific capacitance of 72 F/g at 0.1 A/g was displayed by the PANI/CCNs//PANI/CCNs supercapacitor. PANI/CCNs//PANI/CCNs electrode demonstrated a 28.90 Wh/kg energy density under an 85.10 W/kg power density. Lyu et al. [159] proposed a composite made of yeast-derived N-doped carbon microspheres and

polyaniline (YC/PANI). The active sites for PANI are provided by the wide SSA and hierarchical pore structure of carbon materials made from biomass, and this might lessen the loss of pseudo-capacitance that PANI introduces during charge/discharge. The YC/polyaniline composite, a supercapacitor electrode, provided a three-electrode system with outstanding rate capability and a high specific capacitance of 500 F/g at 1 A/g. On the other hand, the symmetric supercapacitor device made of YC/polyaniline composite demonstrated a high capacitance of 100 F/g at 1 A/g and 95.40% of the original capacitance maintenance after 5,000 cycles. It has become popular to dope heteroatoms and functional materials from biomass precursors, and the activation, as the study of biomass-based supercapacitors moves into deeper territory. The advantages of electrochemical double-layer capacitors and pseudo-capacitors combined by this technique also improve the efficiency of the contact between the electrode material and the electrolyte ions. For the future development of supercapacitors, it represents a significant achievement. However, adding metal and polymer components to charcoal materials is not recommended without drawbacks. Frequent use leads to a decline in the materials cycle stability and ongoing consumption of their pseudo-capacitance, posing challenges to widespread application. Nonetheless, these limitations also highlight key areas for future scientific advancement.

Supercapacitors derived from natural materials such as banyan tree leaves, rose petals, neem tree leaves, and jackfruit seeds have been investigated in several studies. These studies have focused on developing symmetric and asymmetric supercapacitors with improved energy density and electrochemical performance. For example, wood-derived nanostructured electrodes for aqueous asymmetric supercapacitors achieve high areal capacitance and a wide voltage

window [160]. Further, a study reported the fabrication of a high-energy asymmetric supercapacitor using tomato-leaf-derived hierarchical porous activated carbon and electrochemically deposited polyaniline, which showed excellent cyclic stability and efficient operation of a heart-pulse-rate monitor [161]. Hybrid supercapacitors using nitrogen-doped graphene/magnetite and carbon dots/polypyrrole/ $\text{Fe}_3\text{O}_4$ /TEMPO-oxidized cellulose nanofiber electrodes were also investigated, showing higher specific capacitance and energy density [162]. Additionally, asymmetric supercapacitors using manganese-doped Ni sulfide-Sn sulfide/reduced graphene oxide and manganese sulfide/reduced graphene oxide composites as positive and negative electrode materials demonstrated high energy density and cyclic stability [163]. A study confirmed that the tailored hard carbon electrode in carbonate-based electrolytes exhibited significantly enhanced electrochemical performance for sodium-ion hybrid capacitors. The optimised interface enabled a high reversible capacity of  $286 \text{ mAh g}^{-1}$  at  $0.1 \text{ A g}^{-1}$ , with excellent rate capability retaining  $145 \text{ mAh g}^{-1}$  at  $5 \text{ A g}^{-1}$ . Superior cycling stability was demonstrated with 92% capacity retention after 2000 cycles at  $2 \text{ A g}^{-1}$ . The assembled full hybrid capacitor delivered an energy density of  $118 \text{ Wh kg}^{-1}$  at a power density of  $420 \text{ W kg}^{-1}$ , maintaining  $64 \text{ Wh kg}^{-1}$  even at  $5200 \text{ W kg}^{-1}$ , confirming efficient ion transport and stable interface formation [164]. In another study, the layer-by-layer assembled redox wood electrodes demonstrated remarkable electrochemical performance due to enhanced ion pathways and redox-active interfaces. The electrode achieved a high specific capacity of  $312 \text{ mAh g}^{-1}$  at  $0.1 \text{ A g}^{-1}$  and maintained  $178 \text{ mAh g}^{-1}$  at  $2 \text{ A g}^{-1}$ , indicating strong rate capability. Long-term cycling showed 91% capacity retention after 1500 cycles [25]. Further, the nitrogen and phosphorus co-doped mesoporous carbon-graphene nanosheets anode exhibited excellent electrochemical performance for potassium-ion capacitors. It delivered a high reversible capacity of  $312 \text{ mAh g}^{-1}$  at  $0.05 \text{ A g}^{-1}$ , maintaining  $168 \text{ mAh g}^{-1}$  at  $2 \text{ A g}^{-1}$ , showing strong rate capability. Superior cycling stability was achieved with 93% capacity retention over 2000 cycles at  $1 \text{ A g}^{-1}$ . The assembled full device reached an energy density of  $126 \text{ Wh kg}^{-1}$  at  $420 \text{ W kg}^{-1}$ , and still retained  $72 \text{ Wh kg}^{-1}$  at  $5200 \text{ W kg}^{-1}$  [165]. These studies highlight the potential of natural materials for the development of symmetric and asymmetric supercapacitors with enhanced performance. Jackfruit seed-based carbon materials have been investigated for their potential use in supercapacitors. These materials exhibit high surface area and well-defined pore structures, making them suitable for high-rate performance supercapacitors [166]. The specific capacitance of the jackfruit seed-derived carbon materials has been found to be in the range of 208.7 to  $292.2 \text{ F/g}$ , indicating their potential as electrode materials

for high-energy-storage supercapacitor applications [167]. Furthermore, these materials demonstrate excellent cycling stability and high rate capability, with capacitance retention rates of up to 71.6% at high current densities and small capacitance loss even after 10,000 charging-discharging cycles [168]. Overall, the use of jackfruit seed-based carbon materials shows promise for the development of low-cost and scalable nanoporous carbon materials for supercapacitor applications.

## Harnessing green electrolytes in supercapacitors

The utilisation of a mixture of organic and aqueous electrolytes, such as AN,  $\text{H}_2\text{SO}_4$ , and KOH, in the production of biochar from agricultural waste for electrochemical devices is still a highly complicated scenario. In addition to contaminating the soil, this chemical pollutes the ground and surface water. Alternative green electrolytes are now usable in supercapacitors, addressing issues with leakage, packaging, and portability [169]. Green electrolytes will promote the use of organic electronic devices in displays, transistors, and the medicinal industry while also helping to reduce electronic waste [170]. Before green electrolyte's widespread commercialization, the following problems must be addressed: (1) it is crucial to lower the cost of green electrolytes by synthesising them from inexpensive methods, (2) choosing methods that generate minimal toxins and perhaps derived from renewable natural resources and (3) selecting the materials that are readily biodegradable and recyclable for sustainability. Supercapacitors with tremendous energy and power densities are being created with an emphasis on enhancing their electrolyte characteristics, which significantly impact these devices' performance [171]. Ionic liquids, aqueous and organic solvent-based electrolytes, and supercapacitors may all work together [101]. However, there are several drawbacks to most conventional electrolytes. Although ionic solutions could be unaffordable in device applications, organic electrolytes have a poor power density and a relatively high equivalent series resistance (ESR) [172]. The skin toxicity of certain organic electrolytes is more evident [173]. Although ionic liquids are non-flammable and possess low vapor pressure, some hydrophilic types are highly water-soluble. If accidentally released, they could pose environmental risks. Therefore, the liquid form of most electrolytes presents a danger of packaging, shipping, and leakage. To address this issue, substantial research is being done on novel solid and quasi-solid electrolytes in electrochemical devices rather than their liquid equivalents. Polymer electrolytes are a practical substitute for solid electrolytes because they have a conductivity comparable to

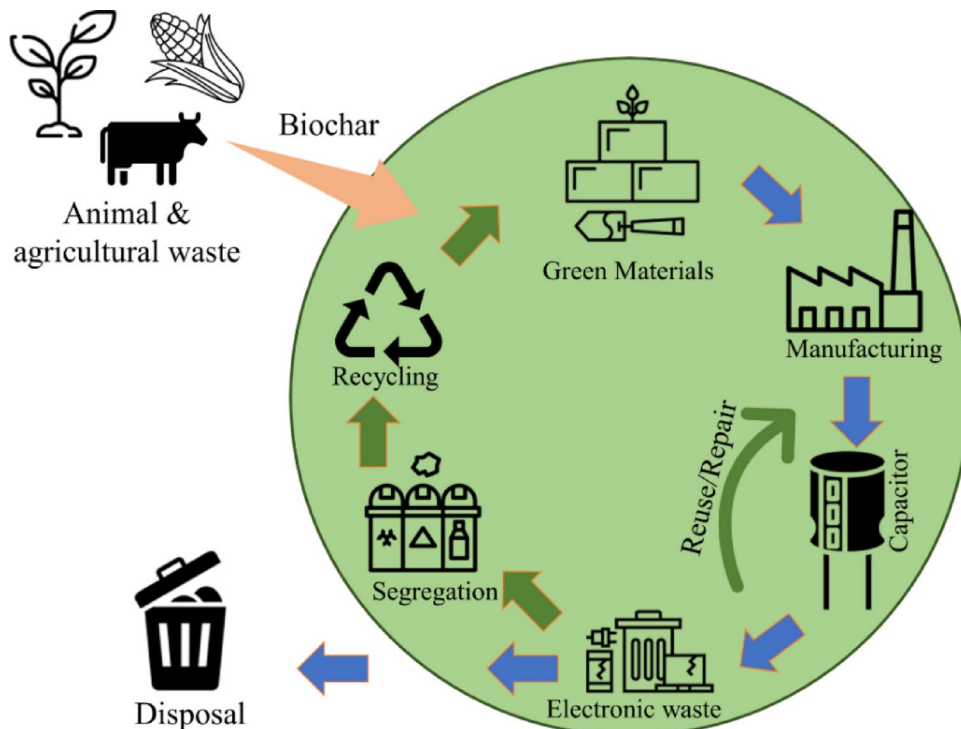
liquid electrolytes and a physical structure similar to solid electrolytes [106]. Studies have shown that different liquid electrolytes may immobilise and become gel-like when applied to solid polymeric or inorganic media. These techniques provide potential supercapacitor electrolyte solutions [174].

## Recycling carbon-based supercapacitor

Recycling electrochemical equipment like batteries has been a common practice in various parts of the world for many years [175, 176]. Here, recovering essential elements and inhibiting objectionable species entering the geosphere are very important. Conventional supercapacitors do not contain valuable materials to be recovered but comprise a substantial quantity of harmful electrolytes scattered in the electrodes [177]. The extraction, processing, transportation, and utilisation of final products and end-of-life products contribute to the circular economy system of agricultural waste-derived nanomaterials. This concept is known as a product's life cycle, and it is presented in Fig. 8. Graphene is frequently utilised to create cutting-edge technology because of its remarkable mechanical and electrical capabilities [178]. Supercapacitors and other electrochemical devices benefit significantly from their use as electrode material. On the other hand, producing graphene in large quantities is expensive [179]. Sepehri et al. [2] developed a wet-shredding process to recover graphene from supercapacitors as supporting fillers for epoxy resins. The

electrodes were cut open and heated to 200 °C to remove acetonitrile. The binder and electrolyte were removed from the electrodes by heating them to 600 °C. The electrolyte and binder may still stay in the electrode to prevent secondary pollution because their presence has little impact on the reinforcing action. The redesigned electrodes were then spread in a ragged fashion in epoxy resin. The HYPSC-002R7-3000 supercapacitor's inexpensively accessible activated carbon was very effective at eliminating dangerous silver and copper ions from water. After the supercapacitor underwent a discharging procedure utilising activated carbon powder, an aluminous current collector, and an organic binder, electrodes were produced from it. The recovered N-methyl-2-carbon had a very high specific surface area of 1403 m<sup>2</sup>/g. After this phase, the binder was removed, and the aluminium foil was peeled off the carbon layer using the many oxygenic pyrrolidone. The carbon material was dried at 600 °C under nitrogen for a whole night to get rid of any leftover binder elements and other metal impurities. Because of this, the activated groups found in activated carbons helped transform hazardous Ag(I) into metallic Ag and reduce damaging Cr(VI) to benign Cr(III). It was discovered that in addition to recovering supercapacitors, filthy wastewater could also be cleaned up [180]. The next step was to construct a fully recyclable EDLC using MWCNT electrodes, MgSO<sub>4</sub> electrolyte, and a di-para-toluene naphthalene diimide (Tol-NDI) organic nanocrystalline dielectric membrane [181]. A simple solution processing method was used to produce and recycle the EDLC. MWCNT and an EP-PDI solution in CHCl<sub>3</sub> were bath-sonicated and filtered

**Fig. 8** Schematic of the concept of the circular economic model aimed at developing sustainable agricultural waste-derived biochar supercapacitors



in a pressure-controlled system to create the electrodes. A Tol-NDI dispersion in oxolane was bath-sonicated to make the membrane. The finished product underwent bath sonication and was diluted down before being aged for five days. Crystals were pressure-controlled, deposited onto a PVDF filtering membrane, dried, and then delaminated to form the film. Before recycling, the membrane and electrodes must be separated. The electrodes were manufactured and then sonicated in EP-PDI chloroform solution, producing a 97% yield. A similar method was used to generate and place the crystals, which resulted in a 90% membrane yield. The structures of the recycled materials matched those of freshly created materials. Studies using cyclic voltammetry on both systems showed exceptional cycling stability after 18,000 cycles. The energy densities of the recycled and new devices were, on average, 0.16 and 0.15 Wh/kg, respectively [181].

Even though supercapacitors have gained attention as a sustainable energy storage technology, their environmental impact when recycled is an important consideration. Recycling waste electrochemical energy storage devices, such as end-of-life supercapacitors, can lead to excellent economic and environmental benefits [182]. By repairing and re-instating the electrode materials from supercapacitors, the structural and surface characteristics of the materials can be maintained, allowing for exceptional cycling stability and high performance in the renovated supercapacitors [183]. Additionally, utilising recycled materials, such as tetrapak/graphene and Ag/BaMoO<sub>4</sub> nanoparticles, in the fabrication of supercapacitors can decrease costs and reduce contamination by plastics in the environment [183, 184]. Furthermore, incorporating solid wastes, such as steel slag powder and waste glass powder, into cement-based materials for supercapacitor cells can enhance their energy storage performances and make them competitive for green building energy storage devices [165]. Thus, recycling and utilising waste materials in the production of supercapacitors can contribute to sustainable practices and the circular economy. Several studies have focused on selecting materials with the least impact on the environment. For example, one study used bio-waste and industrial solid wastes to produce electrode materials for supercapacitors, highlighting the potential of recycling biomass and industrial waste for sustainable energy storage [184]. Another study demonstrated the use of environmentally friendly supercapacitors based on polypyrrole-stabilized polypeptide, which were fully degradable and biocompatible [185]. Additionally, a survey reported active carbon materials from end-of-life supercapacitors. It integrated them into high-voltage and super-stable supercapacitors, emphasising the economic and environmental benefits of waste-recovery initiatives [182]. These studies contribute to the development of eco-friendly supercapacitors and promote sustainable practices

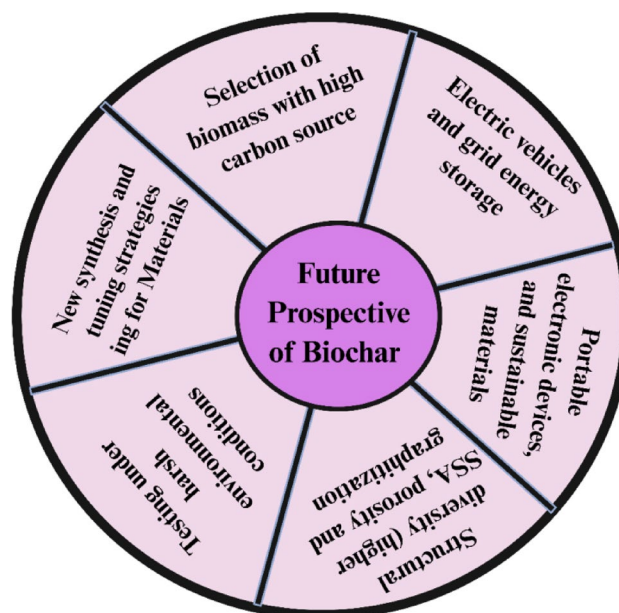
in the circular economy. However, recycling waste materials for supercapacitors has been associated with several drawbacks. One of the main challenges is the need to convert the waste into high-value materials that can be used in energy storage devices. This requires synthetic methods to transform the waste into carbon-based nanomaterials, such as nanofibers, nanotubes, graphene, and mesoporous carbon [186]. Another drawback is the potential contamination caused by microplastics released into the environment when recycling materials containing plastics. This can lead to pollution in oceans and rivers. Additionally, there may be limitations in terms of the electrochemical stability and performance of supercapacitors made from recycled materials. However, research has shown that efficient supercapacitors can be fabricated using recycled materials, such as tetrapak/graphene, which can help decrease the cost of current supercapacitors and reduce plastic contamination in the environment [184].

The decomposition times of supercapacitor materials vary depending on the specific conditions and materials involved. In supercapacitors, the presence of a surfactant has been found to reduce the ageing time by affecting the concentration of degradation products [187]. Further, lithium-ion batteries face challenges in predicting battery lifetime due to trade-offs between energy density, cost, and rate capability [187]. Research on lithium-ion batteries has focused on understanding the physical and chemical processes that contribute to degradation, such as Li-ion mobility and structural damage [188]. The degradation of supercapacitors has been quantified through accelerated ageing tests, with energy cycling causing a more significant decrease in capacitance compared to calendar tests [189]. Elevated temperatures and voltage levels have been found to impact the capacitance and resistance of supercapacitors, with oxidation products and electrolyte salt precipitation affecting performance [190]. The decomposition reaction is a major ageing reaction in supercapacitors, and the production of gas resulting from the electrolyte decomposition reaction was observed [187]. Another study focused on the degradation of commercial supercapacitors at elevated temperatures and found that oxidation products deposited in the positive electrode reduced the capacitance and increased the resistance of the supercapacitors [191]. Additionally, a study on MoO<sub>3</sub> as a supercapacitor material found that the thermal decomposition temperature has an essential influence on the capacitive behaviour and crystallisation of MoO<sub>3</sub> [190]. Finally, simple models for electrochemical supercapacitors were developed to describe the charge-discharge behaviours in the presence of both voltage-independent parallel leakage process and electrochemical decomposition of the solvent [192].

## Future perspectives of biochar-based supercapacitors

The transformation of biomass into valuable materials with various applications is essential for creating a sustainable society and a circular economy. When adopting inexpensive agriculture-based biochar, substantial challenges still need to be solved. With an emphasis on enhancing porosity properties, streamlining processing processes, lowering component prices, and enhancing application performance, work on improving material manufacture proceeded. The following study directions for biochar production and use are recommended in light of the review's findings: Supercapacitors work well with materials obtained from agricultural waste. The benefits of specially created porous materials generated from agricultural waste include (1) low manufacturing costs and high heteroatom concentrations on the planet, (2) well-developed pore architectures and surface chemistry implants can be used to improve anode performance in LIB applications, (3) materials with pores exhibit high electrical conductivity and excellent cycle stability, enhancing supercapacitor performance and (4) adding metal oxide composites, such as NiO, MnO<sub>2</sub>, and Co<sub>3</sub>O<sub>4</sub>, to biochar will improve wettability and electrochemical capacitance. In contrast, introducing heteroatoms (N, S, and P) will enhance SSA (ion-accessible SSA), ion mobility inside the pore network, stability across pH and voltage ranges, and electronic conductivity. In addition, these metal oxides can offer many contact or reaction sites for activities like chemical reactions and adsorption on the surface. Small-scale tests were also conducted to evaluate the technical, financial, and environmental feasibility of the biochar-based supercapacitor use in practical settings. Biochar-based businesses and industrial facilities must be approved to increase the production of highly porous carbon. National policy (such as tax incentives) can also support the production of highly porous carbons from biomass.

Future research in supercapacitors in relation to their combination with renewable energy sources and simulation is an area of interest. As supercapacitors have characteristics such as high power density, fast charge/discharge response, and long life cycles, they are suitable for renewable energy applications. The use of supercapacitors in energy storage systems can assist in power smoothing and provide virtual inertia to the grid, enhancing grid stability. Additionally, the feasibility of using supercapacitors as a form of energy storage on a domestic scale can be explored through simulation, showing potential for replacing electrochemical batteries. Furthermore, the comparison of different supercapacitor models using simulation tools like MATLAB/Simulink can be conducted extensively to assess their behaviours, particularly in terms of charge/discharge cycles. Biochar obtained



**Fig. 9** Future perspectives of biochar in supercapacitors and material applications

from the waste biomass or organic matter with high carbon content offers a renewable and cost-effective source for developing advanced electrode materials. The selection of appropriate biomass is critical to achieving high surface area, porosity, and conductivity essential for energy storage devices. Future research emphasises optimising structural features such as specific surface area (SSA), pore size distribution, and surface functionality to enhance electrochemical performance. The major application areas include electric vehicles, grid energy storage systems, and portable electronics, where rapid charge-discharge cycles and long-term stability are vital (Fig. 9). The use of biochar also aligns with global sustainability goals by utilising agricultural waste and reducing reliance on non-renewable materials. Further developments will focus on novel synthesis techniques, tuning strategies for materials engineering, and expanding the electrochemical window. Additionally, testing under harsh environmental conditions will validate the practical viability of biochar-based supercapacitors. These developments aim to integrate biochar into commercial supercapacitor technologies, driving forward cleaner, more efficient, and environmentally friendly energy systems. Overall, the multifunctionality of biochar makes it a promising material in the advancement of next-generation supercapacitors and circular economy-driven energy solutions.

## Conclusions

Recent developments in the utilisation of biomass as materials for supercapacitors encompass the synthesis and application of carbon composites derived from biomass as electrode materials for supercapacitors, exhibiting exceptional electrochemical performance. Biochar materials with nitrogen doping have also garnered attention as promising electrode materials for supercapacitors due to their renewable nature, significant surface-to-volume ratios, and high electrical conductivity. However, challenges include optimal process conditions, control of ordered pore structure, and scalability. In order to enhance the electrochemical performance, future research must focus on the nitrogen doping types and pore structure. The primary factor influencing the electrochemical properties of carbon materials derived from biomass is determined to be the specific surface area, which plays a critical role in the functioning of electrochemical double-layer capacitors, facilitating the absorption and desorption of electrolyte ions through the provision of active sites. It is posited that the utilisation of specific surface area can be enhanced through the utilisation of biochar materials endowed with mesoporous and hierarchical pore structures, which can potentially expedite the transfer of electrolyte ions. Furthermore, the capacity of electrode materials to generate pseudo-capacitance can be further augmented by the incorporation of heteroatoms and functional groups. It has also been investigated to use biomass-based carbon compounds doped with many heteroatoms, which combine the best electrochemical performance with the concepts of green chemistry. Biomass-based electrolytes have also been studied as a means to enhance the ionic conductivity and eco-friendliness of supercapacitors. Moreover, research efforts have been directed towards the exploration of lignin and cellulose-based supercapacitors, which present economically viable and environmentally sustainable alternatives. These recent advancements in the field of biomass-based supercapacitor materials hold promise for enhanced performance, sustainability, and cost-effectiveness in the realm of energy storage applications.

**Acknowledgements** The author would like to thank the Biomass, Bioenergy and Bioproducts Lab, Department of Chemical Engineering, Manipal Institute of Technology, Manipal, Indian Institute of Technology, GOA, and Guwahati, and Ramaiah Institute of Technology, Bangalore, for providing all the facilities.

**Author contributions** Ranjeet Kumar Mishra: Conceptualisation, data curation, methodology, investigation, visualisation, writing- original draft, and supervision. Sampath Chinnam, D Jaya Prasanna Kumar and Ravi Sankannavar: data curation, investigation, visualisation. Abhishek Sharma and Kaustubha Mohanty: Supervision, administration, and editing.

**Funding** Open access funding provided by Manipal Academy of

Higher Education, Manipal. The authors have no relevant financial or non-financial interests to disclose.

**Data availability** The datasets generated during and/or analysed during the current study are available from the corresponding author upon reasonable request.

## Declarations

**Conflict of interest** The authors announce that they have no known competing financial interests or personal relationships that could have appeared to influence the work presented in this paper.

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