

Review

# Treatment of Biowaste for Electrodes in Energy Storage Applications: A Brief Review

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**Abstract:** Proper and innovative waste management methods still pose a major concern in our present world. Continuous accumulation of biowaste from bio-processing industries, household, organic residues and so on makes the environment polluted and endangers the health of man and other animals. The common waste management methods which include direct dumping into water bodies, open-air combustion, and as land fillers are obsolete and are the major causes of environmental pollution. Conversion of biowastes into valuable materials aids proper waste management, and helps to attain a cleaner environment, in addition to the fact that wastes are turned into wealth. Biowastes are rich in carbon and can serve as excellent precursors for the synthesis of important carbon materials such as activated carbon, graphene, carbon nanotubes etc. Three important methods of converting biowastes into carbon materials are discussed in this review. The electrochemical, adsorption, and electrocatalytic properties of the materials and the applications in electrochemical energy storage devices are also discussed in brief. This review focuses on the synthesis of carbon materials from biowaste residues and their use in developing electrode materials for batteries and supercapacitors. Future perspectives on the need to exploit greener technology for the conversion of biowastes into important carbon materials should be considered.

**Keywords:** environmental pollution; biowastes; carbonization; activated carbon; specific surface area; electrode performance



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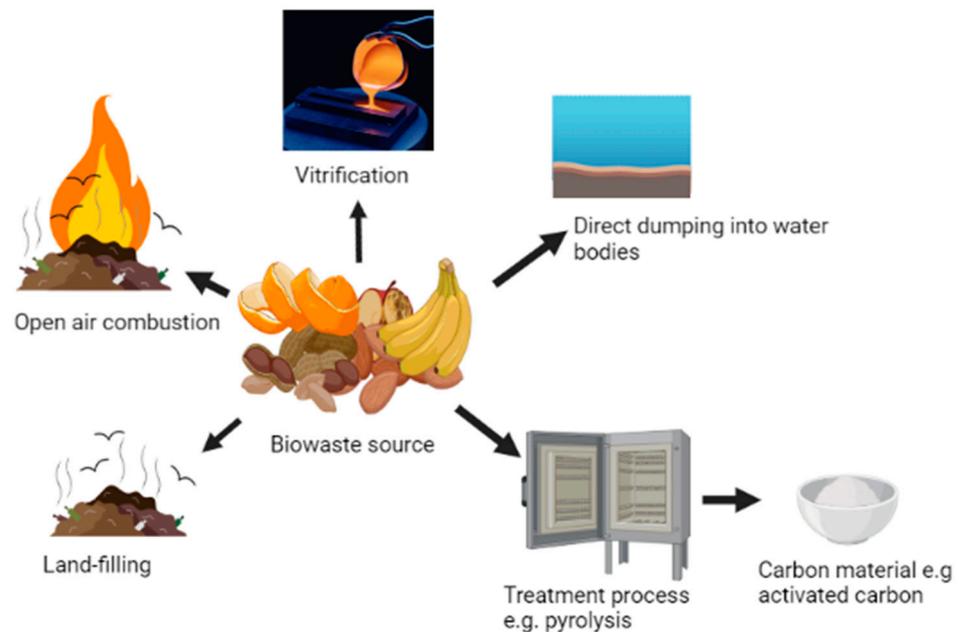
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## 1. Introduction

Biowaste residues are materials that are often generally referred to as waste by individuals, families, and bio-processing industries. The reason is that these materials seem not to be directly useful to them, and therefore are disposed through the easiest available methods which include, direct dumping into water bodies, burning in open air, or as land-filling materials as shown in Figure 1. During the burning and decay of biowaste materials, methane, carbon dioxide, and other toxic gases are generated and released into the environment. These methods of waste management are obsolete and hazardous to the plants, animals, humans, and other non-living components that constitute the environment. Meanwhile, continuous gathering of waste in the environment over a long period of time also pose ecological challenge. Processing of biowastes into useful materials aids proper disposal of wastes which in turn help to attain cleaner environment. Other methods such as vitrification, in which the ash obtained from carbonization of biowastes is subjected to very high temperatures of about 1500 °C to obtain glass-ceramic materials, has been reported as a facile and important method for waste disposal [1–3]. These biowastes such as rice husks, ground nut shells, sugarcane bagasse, spent coffee ground, brewers' spent grains, corncob, coconut shells, wheat husks, and palm kernel shells among others are generated globally in very large amount every year, they are therefore readily available and at no cost [4].



**Figure 1.** Different approach for managing biowaste materials.

For instance, about 700 million tons of rice is produced globally every year, of which nearly 20% by weight comprises husk which is not edible. This amounts to about 140 million tons of rice husk generated per annum [5]. The average amount of waste from groundnut shell is estimated to be between 350 and 400 g per kg of shells produced [6]. Annual global production of sugarcane is estimated to be about 1.6 billion tons, from which about 279 million metric tons of sugarcane bagasse is generated [7]. The industrial processing of coffee is another important source of biowaste to investigate. Spent coffee grounds are recovered as waste after the consumption of coffee. It is estimated that only about 30% of the annual coffee beans produced can be extracted into coffee for consumption, meaning that approximately 70% of the annual coffee beans produced globally are disposed as waste [8]. Similarly, the brewery industry also creates large amount of waste which is economically difficult to deal with [9].

The environmental impact, specifically water and earth pollution, along with the energy demand, is nowadays a serious challenge that face humanity [10,11]. In the case of water, the scarcity and contamination of water sources such as rivers or lakes, are topics of concerning interest, where low-cost technologies such as membranes and adsorbents for metals and other pollutants are stated to be some of the most promising ones [12,13]. On the other hand, developed countries around the world have been recently making efforts to achieve zero-carbon-emissions. This includes minimizing human activities such as combustion of fossil fuels which causes emission of greenhouse gases, promoting the removal of residual greenhouse gases to achieve zero emissions. In 2020, over 100 countries have devoted themselves to taking actions toward achieving zero-carbon-emissions, it is expected that other countries around the world will follow this movement to achieve the global goal [14]. One of the major steps that has been taken toward achieving this goal is to engage researchers by making funds available for research on the conversion of biowastes into important materials for different value-added applications. Variety of treatment processes, such as hydrothermal, pyrolysis, activation, chemical exfoliation, electrochemical, and freeze drying along with others, have being explored to convert biowastes into important materials. These materials find potential applications in energy storage devices, environmental remediation, biosensors, drug delivery, tissue engineering, and so on, either directly or as composites where they are blended with other materials such as metals or polymers.

Different naturally abundant biowastes have been reported for the synthesis of porous carbons due to their availability, high performance, and simple processing methods. Carbon exists in different structural forms, for example activated carbon, carbon nanotubes, aerogels, and graphene which contains large adsorption sites that are important for charge storage and transport. Because of these facts, carbon is an efficient electrode material for batteries and supercapacitors. The various structural forms of carbon that are commercially available for electrode preparation are expensive and not environmentally friendly because they are synthesized from non-renewable sources such as petroleum, coal, and natural gas [15,16]. Therefore, based on the efforts of the developed countries to achieve zero-carbon-emissions in the nearest future, coupled with the race to obtain cheap, abundant, and environmentally friendly carbon in various forms, research attention has in recent years focused on exploring various biowastes as cheap, abundant, and environmentally friendly source of carbon for electrochemical value-added applications.

This review highlights the processing of biowaste materials and their applications in energy storage systems. The review is centered specially in the treatments and preparation of electrode materials from biowaste sources for these applications.

## 2. Biowastes as Rich Source of Carbon Materials

Biowastes are in most cases composed of cellulose, hemicellulose, and lignin which are very rich carbon compounds. Focus on the use of carbon materials from biowaste for sustainable energy development has gained popularity in materials research [15,17–22], it is without doubt because of the high carbon contents of the biowastes as evidently presented in Tables 1 and 2, which make them excellent precursors for the preparation of high-quality materials and nanomaterials. Techniques that have been employed for the conversion of biowastes into carbon materials for different applications include pyrolysis, activation, and hydrothermal treatment among others.

**Table 1.** Ultimate analysis reports of some biowastes.

Biowaste	Carbon (%)	Hydrogen (%)	Nitrogen (%)	Oxygen (%)	Reference
Rice husk	35.82	6.15	5.57	51.95	[23]
Orange peel	45.10	8.78	0.46	42.30	[24]
Coconut shell	42.31	4.65	0.57	52.03	[25]
Sugarcane bagasse	44.60	5.80	0.60	44.50	[26]
Corncob	45.69	6.18	5.65	41.65	[27]
Brewer's spent grain	43.59	6.18	3.46	37.22	[28]
Jute stick	43.41	5.78	7.81	43.00	[29]
Groundnut shell	46.82	6.58	0.80	37.64	[30]
Spent coffee grounds	59.70	7.80	2.20	30.20	[31]
Banana peel	45.43	5.67	2.31	36.40	[32]
Apple pomace	41.70	7.80	0.60	48.10	[33]
Olive mill solid waste	44.10	6.30	1.60	35.80	[33]
Grape seed	47.40	6.70	1.90	38.50	[33]
Grape skin	48.60	7.00	3.00	33.20	[33]
Almond shell	49.62	5.98	0.17	44.23	[34]
Nutshell	48.79	5.99	0.38	44.84	[34]

**Table 2.** Ultimate analysis reports of some biochar.

Biochar Precursor	Carbonization Temperature (°C)	C (%)	H (%)	N (%)	O (%)	Reference
Corncob	1200	89.31	0.45	1.56	-	[33]
Olive mill solid waste	1200	70.76	0.39	2.27	-	[33]
Almond shell	873	85.50	5.28	1.70	7.52	[34]
Nutshell	873	81.13	6.11	1.20	11.56	[34]

## 2.1. Methods for Converting Biowastes into Value-Added Carbon Materials

### 2.1.1. Pyrolysis

Pyrolysis is a process that converts organic matter such as plants or animal residues into carbonaceous materials by subjecting them to high temperatures [35]. Dry biowaste material such as sugarcane bagasse, corncob, coffee shells, rice husk, etc., are exposed to high temperatures of about 700 °C in a furnace in an inert environment (usually nitrogen or argon atmosphere), to evaporate and remove the hydrocarbon contents in an oxygen-free environment. The product obtained from this process is called char or biochar [36]. Bai's group reported the preparation of hierarchical porous carbon from pulp and paper wastes (lignosulphonates) for high-performance supercapacitor electrode via pyrolysis. The pretreated biowaste was pyrolyzed at 900 °C for 2 h in Ar atmosphere followed by ZnCl<sub>2</sub> activation at 900 °C for 3 h [37].

### 2.1.2. Hydrothermal Method

This method is also referred to as high-pressure or high-temperature aqueous carbonization. During hydrothermal treatment, the biowaste is heated at a temperature of about 200 °C in an airtight reactor, usually an autoclave, in the presence of superheated water and self-generated pressure [38]. This process is used to prepare materials with controlled structures [39]. In an experiment, Liu's group synthesized porous carbon from waste coffee grounds for supercapacitor application following hydrothermal synthesis method. They combined catalytic carbonization using FeCl<sub>3</sub> as catalyst with KOH activation at 700 °C, followed by hydrothermal treatment of the resulting carbon material at 120 °C for 6 h to obtain an activated carbon with well-defined porosity (1.64 cm<sup>3</sup> g<sup>-1</sup> pore volume) and ultrahigh specific surface area of 3549 m<sup>2</sup> g<sup>-1</sup> suitable for applications in energy storage devices [40].

### 2.1.3. Activation

Activation of char produced from the pyrolysis process are classified into physical activation and chemical activation. In the physical one, the char is heated at temperature between 800 and 1100 °C in steam, CO<sub>2</sub>, N<sub>2</sub>, or air mixture environment [41]. While in chemical activation, the char produced from pyrolysis is combined and mixed with activating agents such as KOH, NaOH, ZnCl<sub>2</sub>, H<sub>3</sub>PO<sub>4</sub>, K<sub>2</sub>CO<sub>3</sub>, and FeCl<sub>3</sub> in suitable ratios and then heated at a temperature of 800 °C or above to obtain activated carbons with desired properties [42]. Over the past few decades, activation of carbonaceous materials to obtain activated carbon has gained tremendous recognition because of the simplicity, time saving, and cost effectiveness. Activation can be either a one single step or a two-step process which is often preceded by low temperature carbonization or hydrothermal treatment, which helps to control the porosity and surface area of the activated carbon obtained. For instance, Jiang et al., 2020 obtained activated carbon from peanut shell via a two-step activation process. First, hydrothermal carbonization of pulverized peanut shell was carried out at 200 °C for 4 h followed by ZnCl<sub>2</sub> activation at 800 °C for 90 min [43].

## 3. Features of Carbon-Based Electrodes

Two important features that carbon electrode materials should possess are, to be a good electrocatalyst and adsorbent. These features define the applicability of the carbon electrode material in electrochemical energy storage systems.

### 3.1. Electrocatalysis

Electrocatalysis is a process which leads to increasing the speed (rate) of half-cell reactions at the surfaces of electrodes. An electrocatalyst is a catalyst that takes part in electrochemical reactions. They are substances which particularly cause the increase in the speed of half-cell reactions. Electrocatalysts function at electrode surfaces and most commonly may be that electrode surface itself. All-round importance of electrocatalysis in scientific research cannot be overemphasized. Fields such as wastewater treatment,

energy devices, corrosion science, electro-organic synthesis, and development of electro-analysis sensors actively engage the applications of electro-catalysis [44]. The properties of a good electrocatalyst include excellent catalytic activity, high surface area, stability toward anodic corrosion, low energy consumption, and outstanding mechanical and chemical resistance [45].

Electrocatalysts are required to increase the rate of slow electrochemical reactions such as oxygen evolution, oxygen reduction, and hydrogen evolution reactions, which are essential reactions in renewable energy technology. To find alternatives to the existing and expensive metal-based electrocatalysts (e.g., platinum), metal-free heteroatoms such as N, S and P, doped with carbon have been reported as efficient catalysts. A potential electrocatalyst for zinc-air battery was prepared by co-doping porous carbon from garlic stems with nitrogen and sulfur [46]. The electrocatalyst synthesized showed improved oxygen reduction reaction activities with high power density and better stability compared with the platinum/carbon electrocatalyst. Zhang et al. also prepared an electrocatalyst by co-doping nitrogen and phosphorous with graphene oxide. The bifunctional catalyst (i.e., the material prepared acted as both electrode and catalyst) facilitates fast oxygen reduction and hydrogen evolution reactions toward a zinc-air cell [47]. The material is a potential bifunctional catalyst for green energy technology.

### 3.2. Adsorption

Adsorption process involves the movement and gathering of dissolved particles on the surface of an adsorbent material. This process can occur because of physical forces (physisorption) or chemical bonds (chemisorption). It is a reversible process; the reversed process is called desorption [48]. A good adsorbent possesses the following characteristics: excellent capacity, high sorption rate, good selectivity, fast kinetics, and low-cost. The texture, surface morphology, chemical composition of material and chemical activation (functionalization) are important factors which determine the sorption capacity of an adsorbent [48,49]. Physisorption occurs due to weak Van der Waals' attraction forces and because the adsorbent generally has low specific surface area and is non-porous in nature. Meanwhile, chemical adsorption (chemisorption) occurs because of moderately strong chemical connections at the adsorbates-adsorbent interfacial functional groups [48]. The adsorption process is anchored on the physicochemical parameters of the adsorbent material. They include solubility, size of molecule, surface charge, chemical composition, reactivity, and hydrophobicity. Generally, the existence of many functional groups aids the adsorption and storage of electric charges on the surface of the carbon electrode [48,50].

## 4. Biowaste-Based Activated-Carbon and Their Applications in Energy Storage Systems

### 4.1. Batteries

Electrochemical cells can either produce electrical energy from chemical reactions or utilize electrical energy to generate chemical reactions. Batteries are electrochemical cells that utilize reversible redox reactions to produce and store electrical energy. The energy stored in a battery can be used to power electrical devices such as portable electronics (e.g., cell phones) and electric vehicles. Conventional batteries include lithium-ion batteries (LiIBs), lithium-sulfur batteries (LiSBs), sodium-ion batteries (NaIBs), and metal-air (metal-oxygen) batteries [51]. The four basic parts of batteries include cathode, anode, electrolyte, and separator (membrane). Biowastes has been reported to be good precursors for the preparation of electrode materials for battery applications. Table 3 shows the electrochemical performance of some recently reported biowaste-derived activated carbons.

**Table 3.** Electrochemical performance of some reported biowaste-based activated carbon for battery applications.

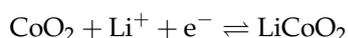
Biowaste Source	Treatment	Specific Capacity (mAh/g)	Coulombic Efficiency (%)	Cycle Number	Application	Reference
Spent coffee grounds	KOH activation	592 at 100 mA/g	97	500	LiIB	[52]
Coconut shell	KOH activation	150 at 0.1 A/g	–	1500	AlIB	[53]
Rice husk/waste coffee grounds	Carbonization	1125 at 100 mA/g	–	100	LiIB	[54]
Corn cob	Carbonization	268.54 at 24.80 mA/g	73.49	–	NaIB	[33]
Grape skin	Carbonization	243.82 at 24.80 mA/g	61.89	–	NaIB	[33]
Grape seed	Carbonization	244.94 at 24.80 mA/g	60.25	–	NaIB	[33]
Apple pomace	Carbonization	247.19 at 24.80 mA/g	40.73	–	NaIB	[33]
Olive mill	Carbonization	174.16 at 28.80 mA/g	50.00	–	NaIB	[33]
Palm kernel shell	KOH activation	869.8 at 200 mA/g	–	100	LiSB	[55]
Rice husk	Carbonization	632 at 1.0 A/g	70.4	100	NaIB	[56]

- i. *Cathode*: Cathode is the positive electrode that acquires electrons from the external circuit. Reduction reactions normally take place at the cathode. Electrons move from the anode into the cathode while conventional current flow out from the cathode. The chemical reactions that occur around the cathode make use of the electrons from the anode.
- ii. *Anode*: Anode is the negative electrode from where electrons flow to the cathode through the external circuit. Conventional current flows into the anode from the cathode. The chemical reactions that occur at the anode-electrolyte interphase causes a build-up of electrons in the anode of a battery. These electrons are prevented by the membrane from moving to the cathode through the electrolyte but move from the anode through the external circuit into the cathode.
- iii. *Electrolyte*: The electrolyte provides the medium for ion transport mechanism between the two electrodes of a cell. Electrolytes could be in aqueous or molten form containing dissolved salts, acids, or alkalis that are necessary for ionic conduction. Nevertheless, some conventional batteries contain solid electrolytes that work as ionic conductors at ambient temperature.
- iv. *Separators*: The separator is a porous material which separates and avoid the anode and cathode from touching each other, which could result in short circuit in the battery. Different materials such as cotton, nylon, polyester, cardboard, and synthetic polymer films are used to prepare the separator. Separators are chemically unreactive toward electrolyte and the electrodes.

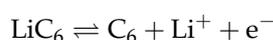
#### 4.2. Lithium-Ion Batteries (LiIBs)

LiIBs are rechargeable battery composed of cells containing lithium ions. When a LiIB is in use (i.e., during discharge), lithium ions ( $\text{Li}^+$ ) move from the anode to the cathode, while during charging, the reverse reaction occurs. Carbon materials are normally used as anode in the conventional lithium-ion cells, the cathode is made of typical metal oxide while the electrolyte composed of lithium salt is dissolved in organic solvent [57]. In a typical LiIB, the following reactions occur during charging and discharging processes [58].

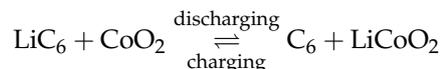
At the positive electrode (cathode) (where lithium is doped with cobalt oxide)



At the negative electrode (anode) (graphite)



The overall cell reaction is



To improve the performance of LiIB, an important aspect to concentrate attention is how to obtain suitable electrode materials, which determine the energy density of batteries. Carbon materials in different forms such as mesoporous carbons, carbon nanotubes, carbon nanofibers, graphene, carbon spheres etc., have been extensively studied and were reported to possess high theoretical specific capacity compared to graphite. This can be seen because of increased storage capacity for lithium ions in the high surface area materials [59].

Interestingly, the synthesis of carbon materials from biowaste residues offer a more convenient and economical alternative. The raw materials are abundant and do not need difficult techniques for treating and processing materials. For instance, in a report by Kwon et al. [60], a composite which was prepared from silicon and carbon material from corn starch was used to fabricate an electrode which was used as anode in a LiIB. The anode presented a capacity of 1800 mAh g<sup>-1</sup> and a capacity retention of 80% after 500 cycles. Moreover, the composite anode was tested in a Li-ion cell using commercial cathodes showing long-term cycling stability at 1 C during 500 cycles. An excellent electrochemical performance of the electrode was recorded which suggests the applicability of corn starch carbon material in LiIBs. In another report [59], graphitic porous carbon sheet with specific surface area of 1246 m<sup>2</sup> g<sup>-1</sup>, synthesized via KOH activation and carbonization of jute stick which was used as anode material in LiIB. This material was reported to demonstrate high specific capacity and outstanding specific capacity retention. These and many other biowaste materials that have been previously studied prove to be promising electrode materials for LiIBs applications.

#### 4.3. Sodium-Ion Batteries (NaIBs)

NaIBs are rechargeable batteries analogous to the LiIBs, but charges are carried by sodium ions (Na<sup>+</sup>) instead of the Li<sup>+</sup>. The working principle and the cell construction of NaIBs are like those of the LiIB types that have gained commercial recognition. A major advantage of NaIBs over LiIBs is the abundance of sodium in nature. Therefore, commercial production of NaIBs is expected to be cheap compared to LiIBs [61].

NaIB cells are made up of cathodes which are based on materials that contain sodium, and the anode is made of inert materials and electrolyte which can be an aqueous or non-aqueous solution of sodium salt. The aqueous electrolytes have the disadvantage of producing low voltage due to the limited electrochemical stability window of water. Therefore, non-aqueous electrolytes such as sodium hexafluorophosphate are often preferred because of their ability to extend the window range [62–64]. During the charging process, sodium ions are removed from the cathode and migrate into the anode where at the same time, electrons travel through the external circuit from the cathode into the anode. The reverse process occurs during discharging. Due to the difference in the physical and electrochemical properties of sodium and lithium, materials generally used for LiIBs are not always suitable for NaIBs [65]. For instance, the graphite anodes used in LiIBs are not suitable for use in NaIBs because it cannot store the sodium ions in significant amount due to the large size of the ions. Rather, non-graphitizable, non-crystalline, amorphous carbon known as hard carbon is presently preferred as ideal sodium-ion anode. Hard carbon has an outstanding cycling stability, high capacity, and lower working potentials. Peanut shells [66], rice husk [61], walnut shells [67], apricot shells [68], apple [69] among others have been reported for the preparation of hard carbon to be use as anode materials in NaIBs applications.

#### 4.4. Other Batteries

In metal-air electrochemical cells, the anode is usually a pure metal with an external cathode of ambient air and an aqueous electrolyte. When the battery is in use, reduction

reaction normally takes place at the air cathode and the anode undergo oxidation [70]. Metal-air batteries possess higher energy density and specific capacity compared to LiBs. Because of this, MABs are becoming important potential candidate for applications in electric vehicles. The commercial application of MABs is faced with complications that are associated with challenges such as slow oxygen-reduction reactions at the cathode which has slow-down its development and commercialization [71].

Introducing electron-rich atoms into the carbon matrix to serve as catalyst for the acceleration of oxygen reduction reactions in air cathodes is found to be an encouraging solution to the problem of slow kinetics of the oxygen reduction reactions in MABs. Ma's group co-doped nitrogen and sulfur in porous carbon derived from the carbonization of garlic stems. The material obtained was used as cathode electrocatalyst with promising faster oxygen reduction reaction capability in a zinc-air cell [46]. In a similar report, heteroatoms such as nitrogen, sulfur, or phosphorus doped with graphene have also shown tremendous catalytic effectiveness toward oxygen reduction reactions [70,72]. Considering cheap, abundant, fast processing and sustainability, heteroatoms doped with carbon from biowaste sources can be utilized as an alternative of graphene-based oxygen reduction reaction electrocatalysts.

Another promising technology is the aqueous Zn-ion battery (ZnIB). In this type of battery, the working principle is the charge transfer between the Zn metal anode and the cathode [73]. They present lower cost and higher safety compared to Li-ion batteries because aqueous electrolytes are much safer than organic electrolytes commonly used for lithium salts. However, their narrow electrochemical window limits their applications and hinders their competence with Li-ion and other technologies. One method to improve their performance is with the addition of a conductive additive to the cathode, as it was reported by Li et al. [74], where they developed a cathode composed of sulfur and carbon nanotubes in a 50 wt.%. They obtained a capacity of 1105 mAh g<sup>-1</sup> and an energy density of 502 Wh kg<sup>-1</sup>, demonstrating the technical and economic feasibility of this type of cathode.

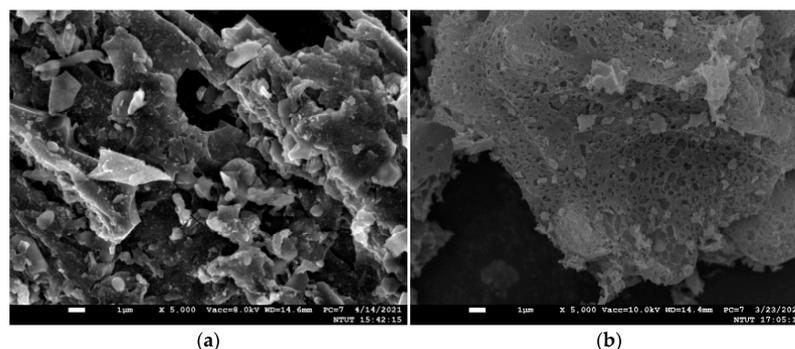
#### 4.5. Supercapacitors

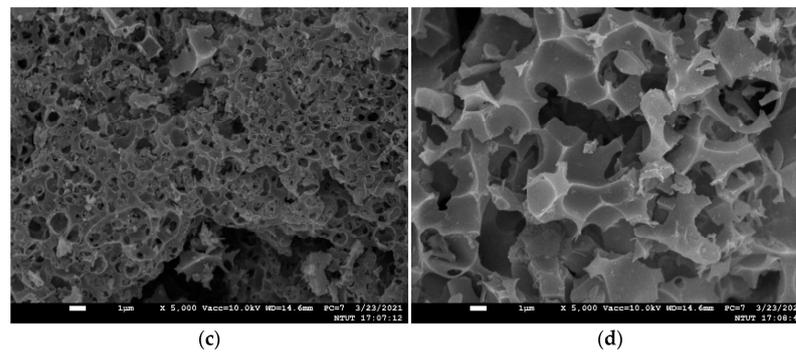
Supercapacitor is another type of energy storage device that stores and gives-out energy at a very fast rate, given the high current within a short time duration [75]. Activated carbon is possibly the most used material in supercapacitors due to its low cost, simple processing method, and high surface area [76,77]. The components and design of the supercapacitors are like those of the batteries. Supercapacitors are made up of electrode, electrolyte, current collector, binder, and separator/selective ion membrane. Although all the components have contributions to the storage performance of a supercapacitor, the electrode and electrolyte both play a major role. In supercapacitors, energy is stored as charges at the electrode–electrolyte interface, the extent of charge storage by the electrode is known as the capacitance. An important parameter to achieve high capacitance is the surface area. Electrodes that possess large surface area have capacity to store more charges and thereby have higher capacitance compared to those with small surface area. Based on the electrode materials, the supercapacitors can be classified into three categories which are: electric double-layer capacitors (EDLC), in which the working principle is based on the Helmholtz layer; pseudocapacitors based on faradaic reactions; and hybrid capacitors which combine both behaviors [78]. The carbon materials such as activated carbon, carbon aerogels, carbon nanotubes (CNTs), graphene etc., show the EDLC behavior. Nevertheless, other alternative materials and structures such as metal organic frameworks derivatives (MOF-D) have demonstrated promising characteristics toward this application [79]. These electrode materials are electrochemically passive, the charge storage takes place only due to the physical buildup of charges/ions on the electrode surface [80]. Table 4 shows the summary of the performances of some reported biowaste-based activated carbon in supercapacitor application.

**Table 4.** Performance of some reported biowaste-based activated carbon for supercapacitor applications.

Biowaste	Treatment Method	Specific Surface Area (m <sup>2</sup> /g)	Energy Density (Wh/Kg)	Power Density (W/Kg)	Specific Capacitance (F/g)	Current Density (A/g)	Reference
Peanut shell	ZnCl <sub>2</sub> activation	2129.5	32.08	1000.0	266.06	0.5	[43]
	KOH activation	3549.0	101	900	224.0	1.0	[40]
Spent coffee grounds	hydrothermal/carbonization KOH activation	1835	10.84	4589	312	0.1 & 3	[52]
Orange peel	CuCO <sub>3</sub> activation	912.4	31.3	499.5	375.7	1.0	[81]
Prosopis juliflora wood	KOH activation	2943.0	56.7	248.8	588.0	0.5	[82]
Corn cob	KOH activation	1722.0	18.84	149.36	382.6	0.5	[83]
Lemon peel	KOH activation	744.78	4.67	8113.0	152.14	-	[84]
Rice hull	NaOH activation	768.0	31.9	309.2	150.8	0.5	[85]
	NaHCO <sub>3</sub> activation	1100.0	20.0	500.0	150.0	1.0	[86]
Spent coffee grounds	CO <sub>2</sub> activation	2497.0	48.3	627.0	222.4	0.5	[87]
Banana stem	ZnCl <sub>2</sub> activation	788.09	6.19	44.67	179.0	-	[88]
Coconut shell	KOH activation	1567.0	48.9	1000.0	449.0	1.0	[89]
Sugarcane bagasse	Activation	1351.7	14.5	125.0	418.5	0.5	[90]

Activation process of biowastes allows to obtain porous materials with a wide range of pore size as it is shown in Figure 2. Lee et al. [89] prepared coconut shell activated carbon using different concentrations of KOH where they reported a superior surface area of KOH-treated samples (1178–1567 m<sup>2</sup> g<sup>-1</sup>) in comparison with the not treated with KOH samples (282 m<sup>2</sup> g<sup>-1</sup>). Liu's group synthesized porous carbon from waste coffee grounds for supercapacitor application following hydrothermal synthesis method. They combined catalytic carbonization using FeCl<sub>3</sub> as catalyst with KOH activation at 700 °C followed by hydrothermal treatment of the resulting carbon material at 120 °C for 6 h to obtain an activated carbon with well-defined porosity (1.64 cm<sup>3</sup> g<sup>-1</sup> pore volume) and ultrahigh specific surface area of 3549 m<sup>2</sup> g<sup>-1</sup> suitable for applications in energy storage devices. The material obtained demonstrated excellent potential toward supercapacitor application with energy density of 101 W h Kg<sup>-1</sup> at 900 W Kg<sup>-1</sup> power density with high capacitance of 440 F g<sup>-1</sup> at 0.5 A g<sup>-1</sup> in 6 M KOH aqueous electrolyte [40]. The porous carbon obtained by Bai's group showed excellent energy storage performance when used to assemble symmetry supercapacitor. It exhibits high-specific capacitance of 289 F g<sup>-1</sup> at a current density of 0.5 A g<sup>-1</sup> with high energy density of 40 W h Kg<sup>-1</sup> at a power density of 900 W Kg<sup>-1</sup> [37]. In another report, activated carbon obtained from peanut shell via chemical activation was found to possess large surface area of 2129.5 m<sup>2</sup> g<sup>-1</sup> with good porosity. Electrochemical analysis revealed that the activated carbon obtained exhibited high specific capacitance of 266.06 F g<sup>-1</sup> at 0.5 A g<sup>-1</sup> current density with energy density of 32.08 W h Kg<sup>-1</sup> at 1000 W Kg<sup>-1</sup> power density [43].

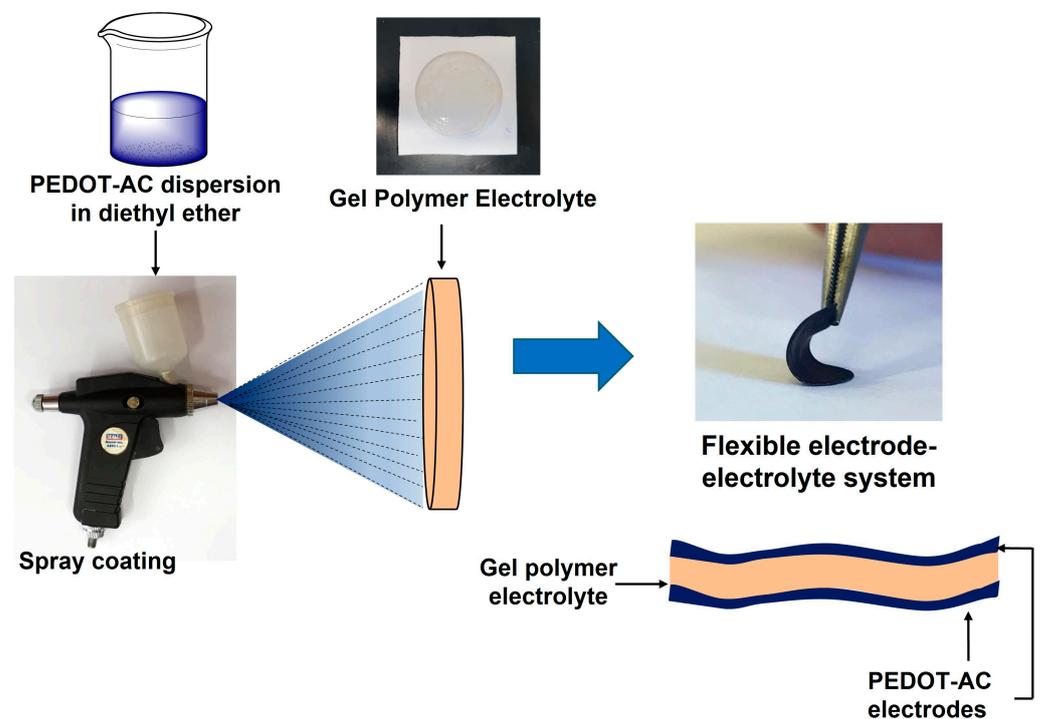
**Figure 2.** Cont.



**Figure 2.** SEM micrographs of coconut-shell derived carbons without KOH (a) and at different KOH concentrations (b–d). Reproduced from MDPI [89].

### 5. Biowaste-Based Composites

Composite materials are prepared to: (i) improve on the performance (such as conductivity and cycle stability) of different metal oxides and conducting polymers such as polyaniline or polythiophenes (Figure 3), (ii) reduce cost, and (iii) reduce environmental hazards. Activated carbon, graphene, carbon nanotubes, and carbon aerogels are the often-used carbon nanomaterials for the preparation of nanocomposites. Carbon nanocomposites offer a high conductivity and specific surface area which is essential for charge adsorption and storage [91]. These carbon nanomaterials can easily be obtained from cheap, abundant, and eco-friendly biowaste sources as discussed in the previous sections. Example of some recently reported biowaste-based composites and their charge storage capacities are presented in Table 5 below.



**Figure 3.** Scheme of preparation of poly(3,4-ethylenedioxythiophene) (PEDOT) and activated carbon derived from brewer's spent grain. Reproduced from MDPI [15].

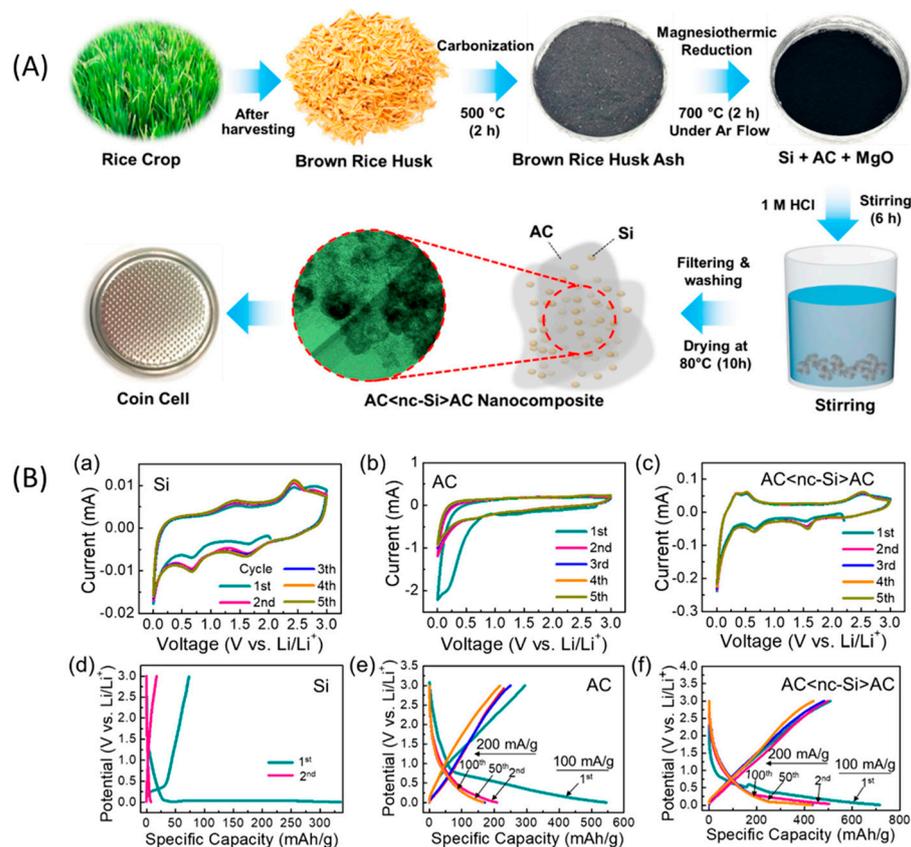
**Table 5.** Performance of biowaste-based nanocomposites.

Biowaste Source	Composite Formed	Electrochemical Performance	Application	Reference
Fennel flower	poly-orthoaminophenol/functionalized graphene oxide/activated fennel flower	1400.2 F/g at 2 A/g	Supercapacitor	[92]
Rice husk	graphene/graphite	16007.02 F/g	Supercapacitor	[93]
Sugarcane bagasse	carbon spheres/graphene	226.8 F/g at 0.5 A/g	Supercapacitor	[94]
Canola waste	poly-orthoaminophenol/functionalized graphene oxide/activated canola waste	1350.5 F/g at 2.0 A/g	Supercapacitor	[95]
Coconut coir	NiFe <sub>2</sub> O <sub>4</sub> /reduced graphene oxide	599.9 F/g at 1.0 A/g	Supercapacitor	[96]
Potato peels	potato peels biochar/copper phthalocyanine	237.0 F/g at 6.1 A/g	Supercapacitor	[97]
Coconut shell	Coconut shell activated carbon/NiO	142.0 F/g at 6 mA/g	Supercapacitor	[98]
Coconut shell	Polyaniline/activated carbon/copper ferrite	248.3 F/g at 1.0 A/g	Supercapacitor	[99]
Wheat husk	NiCo <sub>2</sub> S <sub>4</sub> /wheat husk activated carbon	1962.0 F/g at 1.0 A/g		[100]
Rice husk	P,N-doped carbon/SiO <sub>x</sub>	622 mA/g at 1.0 A.g after 1000 cycles	LiIB	[101]
Rice husk	S,N-doped carbon/SiO <sub>x</sub>	632 mA/g at 1.0 A/g after 1200 cycles	NaIB	[102]
Wheat husk	Sulfur/carbon	822 mA/g at 0.2 C after 100 cycles	NaSB	[103]
Sugarcane bagasse	N,S,O-doped activated carbon	574 mA/g after 1000 cycles	LiIB/LiSB	[104]
Waste tea	Sulfur/activated carbon	428 mA/g at 0.5 C after 100 cycles	LiSB	[105]
Rice husk	Activated carbon/silicon	429 at 100 mA/g after 100 cycles	LiIB	[106]

Continuous effort to improve the electrochemical performance, cost, and environmental hazards is on-going in the materials research community. Very recently, Bigdeloo et al., prepared activated carbon from fennel flower waste as efficient and sustainable source of carbon, the activated carbon was further combined with functionalized graphene oxide to form a binary nanocomposite. A very thin layer of poly-orthoaminophenol was coated on the binary nanocomposite to obtain a ternary nanocomposite of poly-orthoaminophenol/functionalized graphene oxide/activated carbon from fennel flower. Electrochemical measurements were conducted on the ternary nanocomposite material formed using cyclic voltammetry (CV), galvanostatic charge-discharge (GCD), and electrochemical impedance spectroscopy (EIS) to evaluate the energy storage behavior of the electrode material. The ternary nanocomposite shows a specific capacitance of 1400.2 F/g at a current density of 2.0 A g<sup>-1</sup>, capacity retention of 94.4% after 5000 cycles. Brunauer–Emmett–Teller (BET) analysis also reveal that the nanocomposite possesses a specific surface area of 2199.2 m<sup>2</sup> g<sup>-1</sup> [92]. In a similar report, a nanocomposite was prepared from rice husk-activated carbon and silicon nanocrystals by calcination and magnesiothermic reduction process as depicted in Figure 4 (i), to obtain activated carbon-decorated silicon nanocrystals. The composite exhibited an aggregated structure of activated carbon encapsulated nanocrystalline silicon spheres, showing a high specific surface area of 498.5 m<sup>2</sup> g<sup>-1</sup>. The electrochemical performance of the electrode prepared from the composite was tested toward LiIB cell. The cyclic voltamogram of the electrode is given in Figure 3 (ii). The electrode exhibited high coulombic efficiency of 97.5%, discharge capacity of 716 mAh g<sup>-1</sup>, and high reversible specific capacity of 429 mAh g<sup>-1</sup> after 100 cycles [106].

In another report, activated carbon binary composite was prepared from waste olive and date pits, and a ternary composite from olive, date, and sugar. The electrochemical performance of the composites was evaluated and the results showed that the Olive-Date activated carbon composite exhibits a specific capacitance of 104.1 F g<sup>-1</sup> at 0.3 A g<sup>-1</sup> with specific energy of 17.56 Wh Kg<sup>-1</sup>. Meanwhile, the ternary composite (Olive/Date/Sugar) demonstrated better electrochemical performance with specific capacitance of 528 F g<sup>-1</sup> at

$0.3 \text{ A g}^{-1}$  energy density of  $36.82 \text{ Wh Kg}^{-1}$  with high stability after 10,000 charge-discharge cycles at  $10 \text{ A g}^{-1}$  and a capacitance retention of 89.6% [107]. Coconut shell-activated carbon was also used to prepare a ternary hybrid composite with polyaniline and copper ferrite (CuF) via oxidative polymerization. The electrode prepared from the composite demonstrated a specific capacitance of  $248.3 \text{ F g}^{-1}$ , energy density of  $49.66 \text{ Wh Kg}^{-1}$  at  $1.0 \text{ A g}^{-1}$ , and at a maximum power density of  $5996.6 \text{ W Kg}^{-1}$  at  $20 \text{ A g}^{-1}$  in a two-electrode system. It also shows excellent capacitance retention of 92.8% after 2500 cycles which is an indication for a good cycle stability [99].



**Figure 4.** (A) Preparation of activated carbon/silicon nanocrystals composite materials and (B) cyclic voltammetry and galvanostatic charge–discharge (GCD) curves of the materials prepared. Adapted from MDPI [106].

## 6. Conclusions/Future Perspective

Transforming wastes, particularly biowaste materials from bioprocessing industries into useful resources, which can be applied directly or used as raw materials for other industrial processes is a fast-growing research challenge. Biowastes such as rice husk, sugarcane bagasse, nutshell, and wheat husk are rich in carbon and are therefore excellent precursors for the synthesis of varieties of carbon materials such as activated carbon, graphene, carbon nanotubes. Different conversion or synthesis methods such as pyrolysis, activation, and hydrothermal treatments among others have been applied by researchers to convert biowastes into different value-added carbon materials. From recent research reports, carbon materials from biowastes possess high surface area with high porosity which are essential for electrochemical and adsorption performance. Moreover, they can be doped with heteroatoms such as N, P, and S to enhance their electrocatalytic performance. Continuous improvement on the conversion of biowastes into value-added carbon materials will offer a safe and environmentally friendly alternative for waste disposal.

Waste residues such as corn starch, coconut shells, rice husk, coffee grounds, grape seeds, and apple pomace have shown promising properties as precursors for anodes in

LiIB and NaIB technologies, obtaining capacities up to  $1125 \text{ mA h g}^{-1}$  at  $100 \text{ mA g}^{-1}$  for the anode. In addition to this, the preparation of conductive carbon nanomaterials derived from biowaste results as a very appealing approach to improve the performance of the cathodes, in particular for the case of aqueous Zn-S batteries that are a low-cost and safer approach in comparison to other technologies.

Due to their working principle, the conversion of biowaste into high-surface-area carbon materials and nanomaterials seems a promising approach to decrease the cost in supercapacitors, where the treatment used has a direct impact on the surface area and hence in the capacitance and power density of the device. At the same time, when quality electrode materials that can perform excellently in energy storage devices are prepared from cheap biowaste-based carbon materials, the overall cost of the energy storage system will reduce, and it will be affordable by a common man.

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