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Biomass Based Materials in Electrochemical Supercapacitor Applications

Sema Aslan and Derya Bal Altuntaş

Abstract

Biomass is the general term for organic substances derived from living organisms (plants and animals). Since, biomass is a renewable, sustainable, innovative, low cost and carbon-neutral energy source, the applications of nano-micro particles produced from biomass in electrochemical applications have emerged. A large number of carbon-based materials, such as featured activated carbon, carbon nanotube, C-dots, biochar, hybrid carbon-metal/metal oxide ... etc. can be produced from divergent types of biomass. With the growing energy need in the world, supercapacitors have also developed considerably besides the energy generation and storage methods. The supercapacitor is an energy storage system that can work reversibly to provide high energy in a short time. In these systems, electrode structure and surface properties are crucial for energy capacity enhancement. In this sense, electrode modifications with the above-mentioned biomass-based nano-micro structures are widely used in supercapacitor applications.

Keywords: Biomass, carbon, energy, electrochemistry, supercapacitor

1. Introduction

Biomass is defined as all biomass of non-fossil organic matter of biological origin, which can be renewed in less than 100 years, includes land and water-grown plants, animal wastes, food industry, and forestry by-products, and urban wastes [1]. Energy obtained from biomass sources such as vegetable resources, agricultural and animal wastes, organic origin city, and industrial wastes is defined as biomass energy. Since the basis of biomass energy is based on the photosynthesis of plants, biomass energy can also be expressed as the energy of organic matter where solar energy is stored as chemical energy [2].

Vegetable or animal biomass energy sources that can be found in the sea and/or land can be queued as; wood (energy forests, wood residues), oilseed crops (sunflower, rapeseed, soy, safflower, cotton, etc. ...), carbo-hydrate plants (potato, wheat, corn, beet, etc. ...), fiber crops (flax, kenaf, hemp, sorghum, etc. ...), vegetable residues (branches, stalks, straw, roots, bark, etc. ...), animal wastes, urban and cleans [1–3]. Biomass resources are of high water and oxygen content, low density, low calorific value; These features negatively affect the quality of loss [4]. The negative properties of biomass can be eliminated by physical processes (size reduction-crushing and grinding, drying, filtration, aggregation) and transformation

processes (biochemical and thermochemical processes) [5]. Biomass energy stands out with its various advantages such as the ability to be grown almost everywhere, good knowledge of production and conversion technologies, suitability for energy production every time, adequacy of low light intensities, being storable, adequacy of temperatures between 5 and 35°C, being in socioeconomic developments, not creating environmental pollution (Very low NO_x and SO₂ emissions), less greenhouse effect formation compared to other energy sources, cream of CO₂ balance in the atmosphere, not causing acid rain [1–6].

2. Nanomaterials

Studies on nanoscale materials have shown very great improvement and received much attention for decades. Structures are defined as nanoscale materials; Nanocrystals are divided into different classes such as nanoparticles, nanotubes, nanowires, nanorods, or nano-thin films. The main reason for the focus on this subject is that the substances exhibit unusual properties and functionality in a certain size range, unlike their volumetric structures [7, 8]. On the other hand, nanoparticles, which are defined as powders with a size of 100 nm or less, form the basis of nanotechnology due to nanosized materials. These particles show properties that are generally considered different and superior to other commercial materials. The reasons known today for the attractiveness of the frequently mentioned nanoparticle properties are; Quantum size effects stand out as the size dependence of the electronic structure, unique characters of surface atoms and high surface/volume ratio [9].

Nanoparticle synthesis has paved the way for the preparation of many technological and pharmacological products such as high-activity catalysts, special technological materials for optical applications, superconductors, anti-wear additives, surfactants, drug carriers and special diagnostic tools, due to the extraordinary properties these structures exhibit. In addition, the control of materials at the nanoscale level allows the realization of miniaturized devices with specific functions such as nanocarriers, sensors, nanomachines and high density data storage cells [8–10]. The indispensable first step for new developments in nanotechnology, which includes the design, manufacture and functional use of nanostructured materials and devices, is the production of nanoparticles. Nanoparticles, which form the starting point of nanotechnological materials, can be produced in a wide chemical range and morphology. Today, nanoparticles with different morphologies such as core-shell, doped, sandwich, hollow, spherical, rod-like and polyhedral can be prepared from metal, metal alloy, ceramic and polymer-based or mixtures with the desired properties [10–12].

2.1 Biomass based carbon nanomaterials

Biomass is a very low cost carbon source and is used in the production of high value carbon nanomaterials [1–3]. While biochar, activated carbon and mesoporous graphite come first among these materials, when advanced processes are applied, graphene, graphene oxide, carbon nanotubes, carbon-based nanostructures and even hybrid nanostructures containing metaloxide can be successfully produced (**Figure 1**) [13–18].

In particular, biochar, activated carbon and porous carbon production from biomass and their various applications were very popular [16–21]. In one of these studies biochar is synthesized from hair and then activated charcoal without any chemical treatment [19]. The same group reported the synthesis and characterization

Figure 1.
 General flowchart of the green synthesis of high value added carbon materials from the biomass.

of the activated carbon again from waste human hair mass successfully by using chemical activation [20]. Although biochar has a developed porous structure, it shows the limited specific surface area, and activated charcoal has an uncontrolled pore size distribution and irregular three-dimensional structure, which has greatly limited their use in specific applications. For this reason, new generation composite carbon structures originating from biomass were developed, and materials with superior thermal and electrical conductivity and good chemical and environmental stability were developed [22–24]. Later, graphene, graphene-like nano-layers were developed from structures such as activated charcoal obtained from biomass, whose pore structure progressed in a stable pattern with good electrical conductivity, and whose pore structure did not differ [16–22]. Also, Wang et al. [25] reported that the microwave plasma beam method can be utilized to create graphene CNT hybrids.

3. Supercapacitors

The need for energy is increasing day by day and the resources used for energy production are rapidly being depleted. Although the types of resources used in energy production vary, the dominant resources are still petroleum-based fossil fuels [26]. The greenhouse gases released while obtaining energy from these fuels threaten the ecosystem seriously. In this sense, energy storage has gained importance as well as energy production. The methods used for energy storage can be listed as ultracapacitors/supercapacitors, superconducting magnetic energy storage, fuel cells, and batteries [27]. Among these, supercapacitors have an important place. Supercapacitors are also known as secondary batteries used as energy storage and conversion systems. It has attracted great attention in recent years due to its high power density and long cycle life compared to rechargeable batteries [28]. Supercapacitors can be classified as electrical double layer capacitors, pseudocapacitors and hybrid capacitors. The most important factor in improving the performance of supercapacitors is the electrode used [27–29]. Many modifications have been developed to load on the electrode surface and to keep this charge in the double layer for the desired time, that is, to provide charging and discharging when needed. Conductive polymers [30], nanofibers [31], graphene and graphene oxide [32], carbon-based [19] or metal–metal oxide derivative hybrid [33] or nanostructures of different sizes and many other types of modifications.

3.1 Biomass based materials in supercapacitors

Carbon based materials occupy a respectable area on the electrode development in electrochemical energy devices. In general, the source and production methods of carbon-based materials used in batteries, capacitors, supercapacitors and fuel cells have gained importance [34]. The ability to obtain carbon-derived composite or single materials that exhibit superior properties from biomass has made an important contribution to this field. The materials produced to provide high capacitance, fast response and high cycle number, especially in supercapacitor applications [34, 35]. Especially the large surface area due to the porous structure of the carbon-based materials obtained and the ability to be made with an cost-effective and unlimited precursor provides a great advantage [34–42].

Senthil et al. [36] previously examined changes in supercapacitor performance by using the tubular porous carbon material (HT-PC) they produced using waste feather grass flower (FFGF) in electrode modification. They reported a very high specific capacitance value around 300 F g^{-1} with HT-PC containing electrode. Additionally, they obtained % 96 capacitance conservation after 50000 cycles whereas the electrolyte solution is changed from KOH to $1 \text{ M Et}_4\text{NBF}_4/\text{AN}$ electrolyte they observed % 30 capacitance loss over 10000 cycles. This is noted as a very appreciable recovery for capacitance conservation. In another flower based study is reported by Zheng et al. [37]. They used waste-kapok flower as a precursor to produce hierarchically porous carbon as supercapacitor electrode. Here the authors took the advantages of the oxygen rich structure of the kapok flower to obtain micro and meso porous carbon structure. The KOH electrolyte using supercapacitor achieved around 290 F g^{-1} supercapacitance value and showed excellent cyclic stability.

Activated carbons are one of the most popular carbon materials for commercial supercapacitors. Jain et al. [38] reported a cavitation process of the activated carbon from the mixture of native European deciduous trees, Birch, Fagaceae, and *Carpinus betulus* (commonly known as European hornbeam). They reported a desirable carbon with enhanced porosity and high specific surface area of about $614 \text{ m}^2 \text{ g}^{-1}$. Here they examined the supercapacitance of the synthesized activated carbon as an additive to electrode structure in acidic $1.0 \text{ M H}_2\text{SO}_4$ electrolyte. Finally, they showed that the reported method is suitable to achieve a versatile electrode material for supercapacitors.

The hydrothermal method is also a green synthesis method for biomass based productions. Nguyen et al. [39] utilized a carbonization method to synthesize a high-surface-area carbon (HSAC) material from the peanut shells. The HSAC growth on the Ni foam and obtained material achieved nearly two times higher specific surface area than the activated carbon which is produced by Jain et al. Besides they processed the supercapacitor application in KOH and $1 \text{ M Li}_2\text{SO}_4$ electrolyte and obtained a highly desirable capacitance value around 250 F g^{-1} . This is a very unique example of a completely green synthesis of carbon based materials in terms of the source and the processed method. Another impactful biomass based supercapacitance materials are carbon aerogels. They possess highly multifunctional features including compressibility and elasticity. Differently, Long et al. [40] used the calcinated mixture of glucose & dicyandiamide nanosheets (C-GD) and cellulose nanofibers (CNFs) to synthesize nitrogen doped carbon aerogels (C-NGD). C-GD and CNFs lead to a super stable wave-layered structure with an ultimate compression strain (95%) and dedicated as a potential multifunctional material toward flexible electronics, and energy conversion/storage devices.

The porosity of the developed carbon material is very crucial for catalytic applications. To achieve a better regularity in the pore size and distribution divergent

methods are developed. This is very important to obtain consistent activities from the contributed material. In one of these studies, Wu et al. [41] reported that by using melamine foam it is possible to obtain hierarchical porous carbon. They used the waste liquid of the vitamin-C production as the precursor and obtained highly porous carbon by using this template. They used KOH activation on the precursor, and the porous material showed excellent supercapacitance value as 217 F g^{-1} in 1-ethyl-3-methylimidazolium tetrafluoroborate (EMIMBF₄) electrolyte. The cycling stability of the material amendable and the energy and power densities are evaluated as impactful by the authors (**Table 1**). A considerable part of this study is the utilization of the developed material in all-solid state symmetric supercapacitors and obtained attainable capacitance value as 180 F g^{-1} capacitance in gel polymer electrolyte. Because, the multiple applications of the same material enhances their potential and commercial value. Also Fang et al. [42] developed a high-performance flexible supercapacitor which is produced from carbon nanorod and carbon fiber. They used waste straw for the production of carbon nanorods supported hydrothermal carbons and carbon fibers (CNR/HTC/CFs). They reached to 270 F g^{-1} in solid state supercapacitor.

It is possible to apply the produced featured biomass derived carbon materials in both HER and supercapacitors to evaluate their efficiency in energy devices. Cao et al. [52] used the bean sprout to produce nitrogen doped carbon material. Both the carbon and the nitrogen source was the biomass itself. In such a process the control of the pore size and distribution can be controlled by the heat treatment. Here the self-nitrogen doped porous structure evaluated as a good electrolyte ion transferring system for the hydrogen evolution reaction (HER) and supercapacitor applications. They reported very satisfactory HER and specific capacitances and showed that bean sprout have great energy potential for the industrial scale-up with low-cost.

A large amount of the biomass based production methods are covering lignocellulosic biomass residues. Some of the pioneering studies are reported by Selvaraj et al. and Tan et al. Selvaraj et al. [51] produced the activated carbon nanosheets from *Prosopis juliflora* wood carbon waste blocks as carbon precursor. They achieved an ultra-high specific surface area with micro/meso and macro pores and gravimetric capacitances from 400 to 430 F g^{-1} for the different supercapacitance measurement conditions. Tan et al. [8] reported excellent specific surface area and specific capacitance values (**Table 1**) by tuning the initiator biomass material and activation agent. They processed divergent precursors such as cellulose, hemicellulose, and lignin with changing percentages of extractives. It can be clearly seen that the specific surface area enhancement directly brings the increase in specific capacitance value. Yakaboylu et al. [53] reported that the tuning of the pretreatment parameters is very important to control the final pore composition in lignocellulose based activated carbon production. They used the Miscanthus grass biomass as the precursor for the sheet-like activated carbon synthesis and obtained around 190 F g^{-1} capacitance value. The pore size and distribution are controlled by the KOH pretreatment and interconnected micropores are obtained. Thus, the importance of the process control comes out which affects the pore size and volume, cellulose and oxygen amount in the structure, and morphological features which are crucial for the electrochemical adsorption capacity and catalytic performances of produced carbon material.

Sometimes the produced materials can be functionalized by additional groups or heat treatment is applied to enhance the electrical performance. The addition of heteroatoms provides interconnected networks between pores. Chaparro-Garnica et al. [54] synthesized highly porous ($\text{SBET} > 1200 \text{ m}^2 \text{ g}^{-1}$) activated carbon from hemp residue by H_3PO_4 -assisted hydrothermal carbonization (HTC). Then they doped the obtained activated carbon with nitrogen groups and stabilized the

Sample	Biomass sources	Activation	Synthesis method	Surface area (m ² /g)	Morphology	Capacitance	Electrolyte	Ref
C	Fig-fruit	KOH	Pre-oxidation and activation	2337	Highly porous foam-like structure	217 F g ⁻¹ at 20 A g ⁻¹ with three-electrode system	0.5 M H ₂ SO ₄	[43]
N, S-C	Pomelo peel	KOH	Carbonization and activation	2206	Porous structure with micropores	317 F g ⁻¹ at 1.0 A g ⁻¹ with three-electrode system	1 M H ₂ SO ₄	[44]
Co ₃ O ₄ /C	Mollusk shell	—	Carbonization and hydrothermal	—	Cubic Co ₃ O ₄ (1 μm) coated on C (pore diameter about 25 μm)	1307 F g ⁻¹ at 1.0 A g ⁻¹ with three-electrode system	6 M KOH	[45]
NiCo ₂ O ₄ /C	Mollusk shell	—	Carbonization and hydrothermal	—	NiCo ₂ O ₄ nanowires (1.5 μm) grew on Honeycomb-like C	1696 F g ⁻¹ at 1.0 A g ⁻¹ with three-electrode system	2 M KOH	[46]
Ni-Co/N-C	Bacterial cellulose	—	Carbonization and solution co-deposition	405.8	Ultrathin Ni-Co LDH nanosheets uniformly anchored on C nanofibers	1949.5 F g ⁻¹ at 1 A g ⁻¹ with three-electrode system	6 M KOH	[47]
K-C	Banana stem	KOH	Dehydration, porogenic stage and pyrolysis	567	Non-graphitic carbon materials with highly disordered nano-crystalline structure of hard carbon	479.23 F g ⁻¹ 5 A g ⁻¹	6 M KOH	[48]
P-C	Banana stem	H ₃ PO ₄	Dehydration, porogenic stage and pyrolysis	178	Non-graphitic carbon materials with highly disordered nano-crystalline structure of hard carbon	202.11 F g ⁻¹	6 M KOH	[48]
C	Corncoobs	Pyrolysis	Direct pyrolysis	215	Non-graphitic carbon materials with highly disordered nano-crystalline structure of hard carbon	309.81 F g ⁻¹ 5 A g ⁻¹	6 M KOH	[48]
S-C	Potato starch	Pyrolysis	Preheating and carbonization	42	Non-graphitic carbon materials with highly disordered nano-crystalline structure of hard carbon	99.9 F g ⁻¹ 0.3 A g ⁻¹	6 M KOH	[48]

Sample	Biomass sources	Activation	Synthesis method	Surface area (m ² /g)	Morphology	Capacitance	Electrolyte	Ref
C	Walnut shells	KOH	Carbonization and activation	3577	A sheet-like activated carbon with thin-layer pore walls	220 F g ⁻¹ at 100 A g ⁻¹	6 M KOH	[49]
C	Phoenix leaves	Heat and K ₂ FeO ₄	Graphitization and activation	2208	Large micropores (more than 71.8%)	254 and 273 F g ⁻¹ at a current density of 0.5 A g ⁻¹	KOH and H ₂ SO ₄	[50]
C	Waste liquid from the production of vitamin C	KOH	Carbonization and activation	3837	Hierarchical porous carbon	217 F g ⁻¹ at 0.1 A g ⁻¹	EMIMBF 4	[41]
C	Waste liquid from the production of vitamin C	KOH	Carbonization and activation	3837	Hierarchical porous carbon	180 F g ⁻¹ at 0.5 A g ⁻¹	EMIMBF 4 / PVDF-HFP gel polymer electrolyte	[41]
C	<i>Prosopis juliflora</i> wood	Heat and KOH	Carbonization and activation	2943	Rational micro/meso/macro pore size distributed activated carbon nanosheets	588 F g ⁻¹ at 0.5 A g ⁻¹	6 M KOH	[51]
C-K	Cellulose, hemicellulose, lignin	KOH	Carbonization and activation	3135	Hierarchical porous carbon	410.5 F g ⁻¹ at 0.5 A g ⁻¹	6 M KOH	[8]
C-S	Human hair	—	Carbonization	—	Graphene like structure	139.00 F g ⁻¹	6 M KOH	[19]

Table 1.
Properties and the supercapacitor performances of some of the biomass derived carbon based materials.

material with heating. Resulting highly porous activated carbon is applied to a supercapacitor and it showed comparable performances in aqueous and organic electrolytes. In another study, Liang et al. [55] reported nitrogen and sulfur co-doped hierarchical porous carbon (NSPC) with a high gravimetric capacitance around 350 F g^{-1} . They used the $\text{NaHCO}_3/\text{KHCO}_3$ activated foxtail grass seeds as precursor biomass. Also, Cao et al. [52] studied the effect of nitrogen containing groups of NH_4Cl , $(\text{NH}_4)_2\text{CO}_3$ and urea on the electrochemical performances of biomass derived hierarchical porous materials. They reported that, NH_4Cl is proved to be the porogen with the minimum collapse of pollen grain and urea can be identified as the most effective N dopant with the 300 F g^{-1} capacitance value. Du et al. [56] presented a silica activation process for the carbon produced from the carrot biomass. In this study nitrogen enriched porous carbon is produced by a simple activation method. Nitrogen enrichment is preferred to achieve a higher porous structure. Low-cost Na_2SiO_3 served as initiator and provided catalytic effect on the nitrogen doping process. In general the resulting material showed around 270 F g^{-1} . These results showed that the choice of the biomass precursor is very important that, while carrots are used as the carbon source, the vitamins in carrot biomass can serve as a nitrogen reserve. Ariharan et al. [57] reported a facile synthesis of self-phosphorous doped porous carbon material from Honeyvine milkweed (Pod fluff as a precursor) by a simple carbonization route without using any activation process under argon gas atmosphere. They achieved nearly 250 F g^{-1} capacitance value and showed an excellent supercapacitance recovery after 10000 cycles with 95% recovery. They also examined the H_2 storage capacity of the synthesized porous material and achieved successful performances. The reported study offers an effective route for the stable, conductive and highly porous carbon based material synthesis and their effective utilization in both energy applications.

Waste paper cups were used as a source for the synthesis of a carbon support that is loaded with Fluorescein molecules [58]. The resulting composite is successfully applied to a supercapacitor electrode and showed 214 F g^{-1} specific capacitance. Here the main point is that waste lignocellulosic materials are also a very promising tools for electrochemical response enhancement.

Mostly, the regions and their agricultural potentials are determinative for biomass source selection. In China rice based carbon production is very promising and their wastes are also bear huge biomass potentials. Xie et al. [59] used puffed-rice as a precursor material for carbon sheet structures. They applied gradual heat treatment to the source material and achieved the best performance from R-800 sample. They reached nearly 120 F g^{-1} capacitance performance. It is moderate but energy and power densities are recorded as attainable performances. They also utilized the material in microwave absorption beyond a supercapacitor material. So it has been shown that true selection of the biomass precursor can construct a bridge from the biomass derived materials to sustainable development. Orange peels are also used as a biomass source for activated carbon production [60]. The produced activated carbon is indicated as a very highly porous structure and its nanocomposite was produced by the combination with poly aniline. They both utilized for the supercapacitor material. Hybrid structure exhibited nearly 4 folds of higher capacitance value than the natural form. Another citrus based study is reported by Gehrke et al. [61]. The activated carbons synthesized from *Citrus bergamia* peels by activation with phosphoric acid ($\text{AC} - \text{H}_3\text{PO}_4$) and manganese nitrate ($\text{AC} - \text{Mn}_3\text{O}_4$). Among these materials $\text{AC} - \text{Mn}_3\text{O}_4$ exhibited the best electrochemical performance due to the active transition metal content with a specific capacitance value of 290 F g^{-1} . Leaf extracts are widely used for the green synthesis of metallic nanoparticles. In this process the reactive organic groups in the extracts are utilized as reducing and

stabilizing for metals. *Aloe vera* parts are also used for this purpose. Similar to this approach NiO is modified with the biomolecules in the *Aloe vera* extract to enhance the surface properties. Apart from the above mentioned carbonaceous material synthesis based studies here the composite structure is obtained by the modification of the NiO as a metal oxide with the biomolecules. Resulted material improved the anodic and cathodic peak potentials of the NiO and provided longer stability and charge–discharge capacity [62]. He et al. [63] reported an interesting study on the effect of the mixture usage as raw biomass material. The raw biomass composed of, rice husk, reed rod, Platanus fruit, fibers, flax fiber, and walnut Shell. The mixture of these materials is rinsed and grinded than calcinated under nitrogen atmosphere finally Hierarchical porous hollow carbon nanospheres (HCNSs) were fabricated. This one step process is also performed by the addition of polytetrafluoroethylene (PTFE) to raw biomass. Both the hollow carbons served well as a supercapacitor additive since they possess core-shell pores. Also silica content improved the mesoporosity of the structure very much.

4. Conclusion

The utilization of the carbon based materials in the improvement of the electrochemical performances of energy production and storage devices has reached an important stage. In this manner, the source depletion for the synthesis of these materials leads the researchers to find new and cost-effective solutions. Today's studies show that biomass provides a real ocean to overcome this problem with many advantages. Especially supercapacitors need reliable modifications in which biomass derived carbon based materials play a crucial role. After extensive investigations biomass is found to be capable of the synthesis of highly porous carbon materials with low/no-cost and eternal precursor supplementary. At this point it has to be underlined that these methods not only provide a way to produce high-value added materials but also contribute to the recycling of the wastes with the win-win principle. The reported studies prove the developed biomass derived materials enhance electrochemical adhesion of the ions which leads to increased specific capacitance of the electrode, consecutively cycling ability and stability of the supercapacitor is enhanced. The crucial points in the biomass derived production are indicated as the choice of the biomass, synthesis process, pretreatment, and the type of the supercapacitor. Among them precursor material and pretreatment play a key role because the pore size and distribution vary very much depending on the precursor content and the pretreatment process. Heteroatom doping to the biomass derived materials add extremely high conductivity to the ordinary materials so the composite materials are preferred in many supercapacitor applications. However, because of the biomass derived synthesis is a green synthesis method the researchers avoid to exaggerated hazardous chemical pretreatments, instead they should choose the right precursor biomass material that has this feature in itself that is reported in this study as well. Overall, biomass is a very valuable source for the synthesis of the next generation of low-cost and green electrode materials for supercapacitors, fuel cells, batteries, and all electrochemical transducers.

Conflict of interest

The authors declare no conflict of interest.

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