

## **RECENT TRENDS IN NON-FARADAIC SUPERCAPACITOR ELECTRODE MATERIALS**

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### **Abstract**

Global demand for energy is on a progressive increase and there is a need for environmentally friendly technologies to meet this demand. Electrochemical energy systems are hinged on clean and sustainable technologies. The latest trend in electrochemical energy systems is the supercapacitors (SCs). SCs are famous for their attractive properties: power density, charging time, life cycle, operational safety, and simplicity. However, their energy density is generally low and to a great extent, this parameter is invariably dependent on the nature of electrode material used. While high energy density is being sought for in SCs, it is necessary to keep abreast of recent electrode materials and their practical performances. This paper gives a concise description of capacitors with a focus on the non-Faradaic SCs. It also compendiously presents an overview of carbon electrode materials with their practical performances (specific surface area, specific capacitance, energy and power densities) for non-Faradaic SCs, with reference to more than 100 reputable works. Development and investigation of highly active carbon materials with optimized electrolytic compatibilities and manipulative morphologies and pore structures were recommended.

**Keywords:** ultracapacitors; EDLC; carbon electrode materials; energy storage; Helmholtz double layer.

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## Introduction

Progress in global industrialization alongside world population growth suggests that there would never be a slackening in global energy demand. Although several sources of energy are in place today, inadequate energy storage techniques and continuous depletion of non-renewable and non-sustainable fossil reserves coupled with the accompanying environmental pollution, call for sustainable and clean energy sources with efficient technologies for conversion and storage. Electrochemical energy conversion and storage systems which include secondary cells, supercapacitors (SCs) and fuel cells have proven to be one of the most effective, sustainable and eco-friendly panacea in this regard [1–4].

Electrochemical capacitors, also known as supercapacitors or ultracapacitors, are an advanced form of electrolytic capacitors. They possess some desirably salient features like high power density, long charge/discharge cycle, short charging time, a wide range of operating temperature, operational safety, and simplicity [2, 5]. Considering the power and energy densities, they span from dielectric capacitors to secondary cells [6]. SCs have significantly caught the attention of researchers as evinced by numerous works published on this subject. Fig 1 presents the statistics of publications on SCs available from Elsevier's database (Scopus indexed publications) as at 23<sup>rd</sup> May 2019. The publications include journal articles (19,004), conference articles (3,920), articles in press (accepted manuscripts, 302), book chapters (162), dissertations (147), books (15), errata (3) and a standard. It is evident from the bar chart that research interest in this area is increasing steadily. Meanwhile, an 'all-time' search on Google Scholar reveals approximately 221,000 and 196,000 publications, with and without patents, respectively. Nonetheless, SCs are quite expensive with relatively low energy density [5, 7].

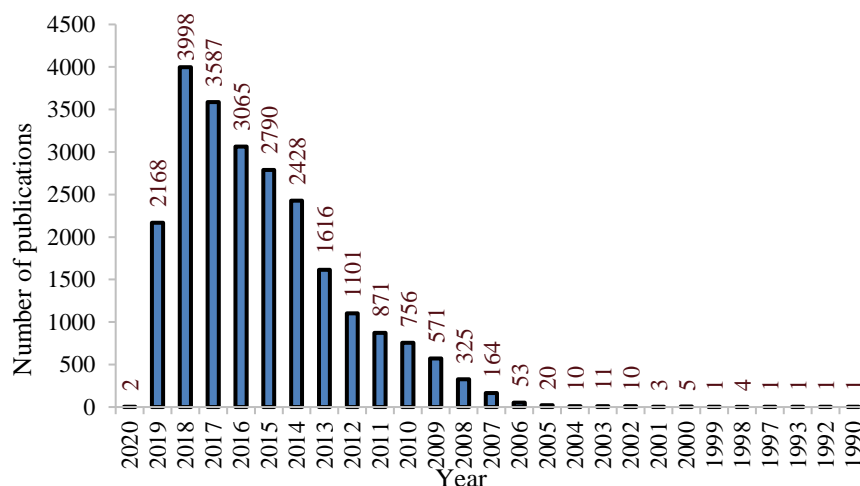


Fig. 1. Statistics of Scopus indexed publications on SCs as at 23<sup>rd</sup> May 2019 (provided by Engineering Village ©2019 Reed Elsevier).

Consequently, most studies have been geared towards improving the energy density of SCs. The expression for energy density ( $E$ ) is as shown in Eq. 1, where:  $C$  is the capacitance of the SC and  $V$  is the potential drop across the double layers [2, 7]. It follows, therefore, that any effective attempt to increase the double layer capacitance or voltage would improve the energy density. Wu and Cao [7] utilized a schematic illustration to show that capacitance is affected by surface area, pore size, electrical conductivity, and available functional groups. Based on theoretical models, Yu *et al.* [2] noted that double layer capacitance of an SC is dependent on the quantity of charge, temperature, the concentration of electrolyte and solvent used; however, they went further to show that the nature of electrolyte and electrode used are of ultimate relevance.

$$E = \frac{1}{2} CV^2 \quad 1$$

Since the choice of electrode and electrolyte are of paramount importance for the performance of SCs, this paper reviews recent researches that employed carbon electrodes for SCs. In other words, only the pure or true electrostatic supercapacitors – those that do not exhibit Faradaic reactions – are covered here. For SC electrolytes, refer to the work of Zhong *et al.* [8].

## Advancement from Capacitors to SCs

### Ordinary Capacitors

A capacitor is a passive electric charge storage device comprising of two close conductors (electrodes) which are electrically insulated from each other. The insulating dielectric is usually sandwiched between the electrodes. Although the knowledge of static electricity has been since 600 BC; courtesy of Thales of Miletus [9], it was not until 1745 that the German bishop and scientist, Ewald Georg von Kleist invented the first capacitor (then known as condenser) after an unusual shock in an attempt to convey and store

charges in a handheld glass jar filled with water [10]. After some months, Pieter van Musschenbroek, a Dutch physicist at the University of Leiden, coincidentally and independently, after a similar experience that made him write to French Mr. Reaumur, "I would not take a second shock for the kingdom of France", invented a similar capacitor which is now known as the Leyden Jar [11, 12]. Thereafter, Benjamin Franklin simplified the design using flat glass piece as dielectric [12]. Fig 2 shows these earlier designs of the capacitor. Today, there are varieties of capacitor designs with variations in electrode geometry (parallel plates, co-axial cylinders, concentric spheres, and isolated sphere) and dielectric material (vacuum, non-ionized gases, insulating solids and liquids). Actually, the dielectric is the active charge storage component of a capacitor, as inferred by *Franklin* [13].

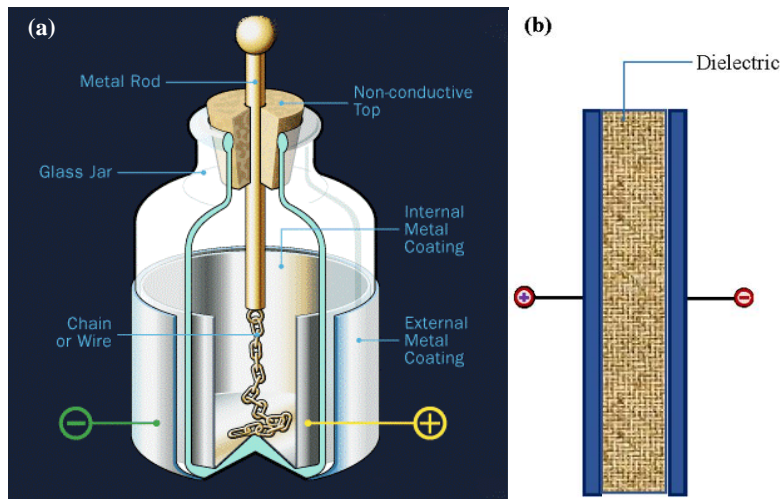


Fig. 2. Earlier designs of the capacitor: (a)- the Leyden jar [12] and (b)- parallel plate capacitor.

A measure of the amount of charge needed to induce a potential of 1 Volt across the electrodes of a capacitor is called the capacitance of the capacitor. For linear capacitors, this property is independent of the quantity of charge nor the potential difference, but upon the geometry of the device and the nature of dielectric used (permittivity,  $\epsilon$ ); larger electrode surface area ( $A$ ) and shorter separation between charged layers or inter-electrode spacing ( $d$ ) improve capacitance ( $C$ ) as shown in Eq 2. Michael Faraday, an English chemist, demonstrated the first practical capacitor and how to use it for electric charge storage in his experiments. Faraday introduced a means of measuring the quantity of charge that can be held in a capacitor and this was measured in Farads, which is equivalent to Coulombs per Volt [12]. When the terminals of a capacitor are connected to a direct current source (charging mode), the source provides an electromotive force (emf) that drives electrons from the positive capacitor terminal to the positive source terminal, while simultaneously feeding electrons to the negative capacitor electrode at the same rate. Charging continues until the potential between the capacitor electrodes equates that of the emf source; then, the capacitor is fully charged concerning the driving potential [2]. During the discharge process, electrons flow in a reverse manner

through a connected load; of course, the flow of electrons implies a flow of current but in reverse direction.

$$C = \frac{\epsilon A}{d} \quad 2$$

In 1896, Karol Franciszek (Charles) Pollak, a manufacturer of secondary cells filed a German patent (GB189601069) for an electric liquid capacitor with aluminum electrodes, after he discovered that the oxide layer on an aluminum anode remained stable in a neutral or alkaline electrolytic system after the circuit was opened [14]. He described his invention as a "liquid condenser with aluminum electrodes, which are covered with a uniformly insulating layer generated by forming with a weak current, characterized by using alkaline or neutral electrolyte. Since the insulating layer is very thin, the condenser has a very high capacitance and could be used as a polarized capacitor in a DC circuit." [15]. Therefore, electrolytic capacitors have thin insulating oxide films as their dielectric and they offer greater capacitance than the earlier types.

Capacitors are the most used electronic component after resistors. They are relevant in several applications, for instance in digital (electronic) circuits for information back-up, so that information stored in computer memories is not lost upon electric power failure; the stored energy in such capacitors maintains the information during temporary loss of power. Capacitors play an even more important role as filters for diversion of spurious electric signals, thereby preventing possible damage by electric surges to sensitive components of electric circuits, a typical example can be found in induction coils.

#### *Concept of Helmholtz or Electric Double Layer (EDL)*

When an insoluble charged or electrically active body is introduced into an electrolyte, there is to a certain degree, an alignment of the mobile ions (of the electrolyte) concerning the new charged surface. Electric Double Layer (EDL) is "a region existing at the boundary of two phases and assumed to consist of two oppositely charged layers (such as a layer of negative ions adsorbed on colloidal particles that attracts a layer of positive ions in the surrounding electrolytic solution)" [16]. Hermann von Helmholtz first realized this concept. He observed that a charged electrode immersed in electrolyte repels the co-ions while attracting the counterions to their surfaces; thereby forming 2 (double) layers of opposite polarity at the electrode-electrolyte interface [17]. This phenomenon was not recognized until 1879 [18]. The EDL is also referred to as the Helmholtz double layer. Helmholtz's concept has been developed successively by Gouy and Chapman; Stern; Grahame [2]; Bockris, Devanathan and Müller [19, 20]; and Conway [21].

Fig 3 is an illustration of double layer formation at the interface of negatively charged particle/rod and electrolyte. Static charges reside on the outer surface of materials, with higher concentrations at sharp edges. When introduced into the electrolyte, the material's charge distribution remains, while the mobile ions of the electrolyte get attracted (counterions) or repelled (co-ions) but the distribution of ions is somewhat diffuse – not as ideal as the material's surface charge distribution. A negatively charged insoluble material in an electrolyte attracts positively charged ions to its neighborhood to form an EDL. Electric double layers of negative (on the material surface) and diffuse positive charges can be seen in Fig 3; the layers are demarcated with a green

dash line. The interlayer spacing is in the order of 1 Å and more detailed description of this concept is given by Yu *et al.* [2], Park and Seo [22].

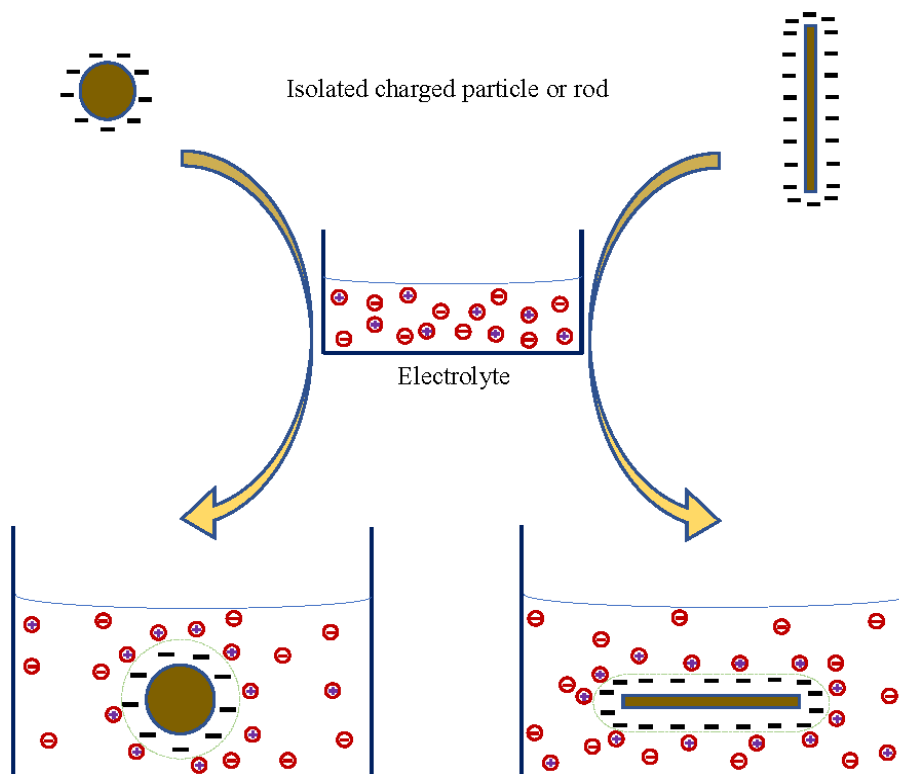


Fig. 3. Double layer formation at the interface of negatively charged particle/rod and electrolyte.

#### Ultracapacitors or Supercapacitors (SCs)

These are advanced or current generation capacitors with capacitance values much higher than a conventional capacitor. SCs can store 10 to 100 times Joules of energy per unit mass or volume when compared to the conventional capacitors [1]. While the ordinary (earlier generation) capacitors were developed for primary circuit applications, with storage of picofarad to microfarad charges in direct current circuits and filtration of frequencies in alternating current circuits, the current generation of capacitors, the SCs, are capable of serving more sophisticated purposes. Historical background of SCs can be found in the work of Samantara and Ratha [18].

A typical SC has two highly porous electrodes on a metallic collecting plate, electrolyte and an insulating membrane or separator between the electrodes that enables permeation of electrolyte ions but prevents contact/short-circuiting. The arrangement is similar to that of a secondary cell. Now, what makes SCs 'super' over ordinary capacitors? The SC electrodes are of extensive surface area in the order of  $10^3 \text{ m}^2\text{g}^{-1}$  and the charge separation is typical of an EDL as earlier stated. Relating these unique features with Eq. 1 and 2, the prefix 'super' or 'ultra' would be found deserving of these advanced

capacitors [23]. The fact that charge separation in SCs follows the EDL principle, has fetched SCs the name Electric Double Layer Capacitors (EDLCs). Fig 4 is a schematic diagram of a supercapacitor revealing the EDL at the non-intercalated electrolyte-electrode interface and the separation between charged layers ( $d$ ) in reference to Eq. 2. Since there is no permeation across the EDL interface (no charge intercalation), the tiny interface can be regarded as the dielectric for EDLCs but actually, they can be permeated as we would see in the case of pseudo-capacitors. The collectors are metallic plates onto which a porous substance (electrode material) such as activated charcoal or graphene nanoparticles is deposited. The large BET surface area of the electrode material offers SCs much more charge storage sites in an ideal situation where the electrolyte wets all the sites. However, in reality, all pores are never accessible [2, 8, 24].

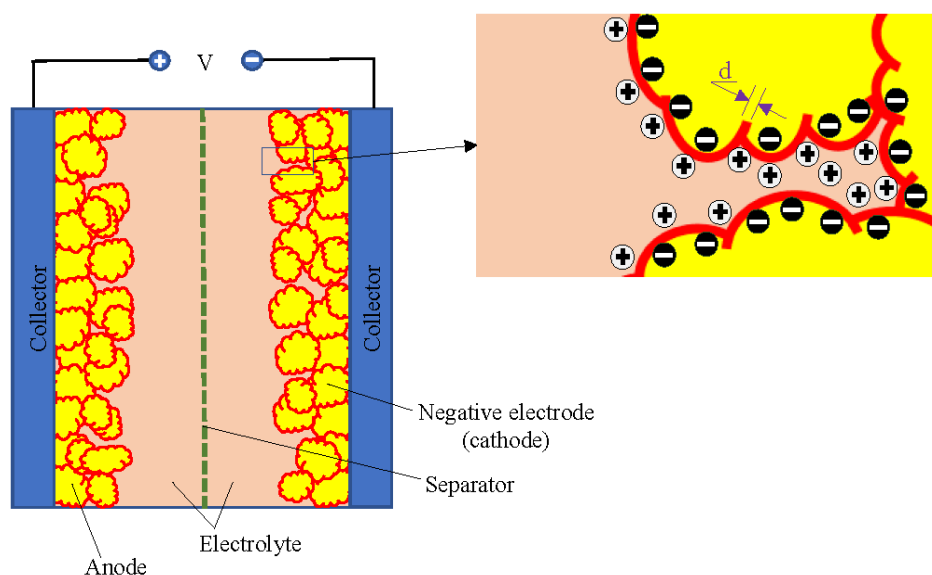


Fig. 4. Schematic diagram of a supercapacitor revealing the EDL formed at the electrolyte-electrode interface and the separation between charged layers ( $d$ ) in reference to Eq. 2.

Furthermore, the mechanism of operation of SCs is best described alongside their classification. First, the 'true supercapacitors' store energy via physical adsorption of ions based on the EDL concept when an emf source is applied. Hence, charge storage is purely by electrostatic accumulation like in ordinary capacitors; non-Faradaic mechanism – no occurrence of a redox reaction. Consequently, this class of SCs are called non-Faradaic capacitors, electrostatic supercapacitors or even supercapacitors/EDLCs (since they are the true ones). This class of capacitors employ carbon electrodes and are characterized by relatively rapid charging/discharging, longer life cycle, and greater coulombic efficiencies than others [2, 5, 24–26]. Their merits come from the fact that the electrostatic mechanism is very much reversible and instantaneous.

Secondly, we have pseudocapacitors. They are the Faradaic capacitors, redox capacitors or the true electrochemical supercapacitors. The mechanism of

pseudocapacitive charge storage is quite different from that of EDLCs. Here electrochemically active electrode (usually composed of transition metal oxide) material is used to bring about redox or Faradaic reaction. When connected to a source of emf, there is adsorption of ions, followed by fast and reversible redox reactions between the electrode materials and electrolyte, on the electrode interface. This reaction involves the intercalation or passage of charges across the EDL, resulting in a flow of Faradaic current, like the charging/discharging of secondary cells. Electrode materials for this class of supercapacitors include transition metal oxides and conducting polymers (such as polyaniline and polypyrrole) [2, 4, 21, 27, 28]. With conductive polymer-based electrodes, there is reversible electrochemical doping/de-doping [27]. Pseudocapacitors offer larger capacitance values and energy density than EDLCs, but they lag in power density due to the relatively slower Faradaic processes involved – reactions have to occur with intercalation of ions. They also suffer lower life cycle due to wear and tear from repeated Faradaic processes.

Lastly, hybrid supercapacitors are those formed by combining certain aspects of EDLCs with those of pseudocapacitors. *Jain et al.* [29] report the onset of Faradaic reaction in carbon electrode SC system with the introduction of p-hydroxyaniline to 1 M aqueous sulphuric-acid electrolyte. This could be a lead to designing hybrid SCs with carbon electrodes using mixed electrolytes. *Zhong et al.* [8] utilized a schematic diagram to show these classes of a capacitor and they noted lithium ion capacitor as a typical hybrid SC. A detailed comparison between the Faradaic and non-Faradaic supercapacitors, as well as their contrasting features with secondary cells are covered in a technical paper by *Conway* [21].

Although SCs boast of high energy storage capacity compared to ordinary capacitors, they still lag behind batteries in that regard. Also, they are usually more expensive per unit than batteries [5, 30]. On a technical note, it is possible to replace a cell phone battery with an SC, and it will charge much faster, but it can only stay charged for a while. SCs are very good at acquiring or delivering a sudden surge of energy and this makes them right candidate for hybrid energy systems with primary energy sources like the internal combustion engines, secondary and fuel cells which are capable of producing energy continuously but at low power. For instance, in powering an electric car, an SC can provide the power for acceleration, while a battery provides over the range of uniform motion and recharges the SC in-between surges [30].

### Electrode Materials for Non-Faradaic SCs

Carbon-based materials have some salient features such as high chemical stability, structural polymorphism, low cost, wide potential windows, relatively inert electrochemistry, rich surface chemistry or electro-catalytic activities for a variety of redox reactions [31–33]. For this versatile nature, varieties of carbonaceous materials have been developed and studied extensively for several electrochemical applications; more importantly, for energy generation/conversion and storage [31, 34, 35]. Use of graphene-based materials in electrochemistry has been reviewed by *Chen et al.* [36].

Electrode materials for SCs are desired to be highly porous with electrolyte-accessible pores, electrically conducting and cheap. More specifically, non-Faradaic SC electrodes are desired to be electrochemically inert and this makes carbon materials a perfect candidate. Techniques for synthesis of non-Faradaic SC electrode materials include carbonization of biomaterials, followed by surface enhancement via thermal



or/and CA, chemical vapor deposition [35, 37], electrospinning [28], electrodeposition of electrode films via an electrolytic coating, direct coating, etc. [38]. In order to create abundant micro-, meso- or macro-pores within the carbon material, soft and/or hard templates are usually employed in the carbonization process, after which they are removed without further chemical/thermal activation (CA or TA). This is known as template carbonization method [39]. Methods of electrode performance assessment are described in the review work of *Stoller and Ruoff* [40].

Finally, we present some trending carbon-based materials that have been tried as electrodes in supercapacitors with their practical performances: specific surface area, capacitance, energy and power. In the consolidation of our earlier review [35], this work reveals one of the unlimited potentials of carbon. Review on this topic has been done before [3, 38, 41–43], but for the first time, we compose concisely, practical performance results of numerous carbon electrodes for non-Faradaic SCs. Table 1 shows the performance of carbon electrode materials in SCs.

Table 1. Performance of carbon electrode materials in SCs in order of decreasing super-capacitance.

Carbon Source	Electrode type	Method of preparation	Charge/discharge cycle	Specific Capacitance /F g <sup>-1</sup>	Current density /A g <sup>-1</sup>	Electrolyte	Surface area /m <sup>2</sup> g <sup>-1</sup>	Energy Density /Whkg <sup>-1</sup>	Power Density /W kg <sup>-1</sup>	Ref.
Wet-blue leather solid waste	Porous carbon	Carbonization and CA	10,000	2203	—	1 M Na <sub>2</sub> SO <sub>4</sub>	—	624.7	749.9	[44]
Wet blue leather solid waste	Porous carbon	Carbonization and CA	500	1833	—	1 M H <sub>2</sub> SO <sub>4</sub>	—	—	—	[45]
Flower	AC	Carbonization and CA	5,000	711	1	1 M KOH	1098	—	—	[46]
Prawn shell	N-doped AC	Simultaneous carbonization and CA	5,000	695	0.05	1 M H <sub>2</sub> SO <sub>4</sub>	—	—	—	[47]
Mango kernel	AC	Carbonization, pulverization and CA	5,000	587.1	—	P-hydroxyaniline in sulphuric acid	—	34.3	—	[29]
<i>Rhus typhina</i> fruits	N-doped porous carbon	Pyrolysis and CA	10,000	568	1	1 M H <sub>2</sub> SO <sub>4</sub>	2675	—	—	[48]
Yeast	N-doped YC/PANI	In-situ polymerization	5,000	500	1	1 M H <sub>2</sub> SO <sub>4</sub>	—	—	—	[49]

Ethanol and cotton wool	CNTs on cellulose derived carbon	Chemical vapor deposition	—	484.34	2	1 M H <sub>2</sub> SO <sub>4</sub>	—	46.34	830	[50]
<i>Rhuspyrhina</i> fruits	Porous carbon	Pyrolysis and, CA	10,000	474	1	6 MKOH	2675	—	—	[48]
Naphthalene	ACFs	CA and carbonization	1,000	427	0.1	6 MKOH	—	—	—	[51]
MRF	NGHPC	Carbonization, graphitization and CA	—	417.5	0.5	6 MKOH	2729	—	—	[52]
Pea protein	N and O dual-doped porous carbons	Pyrolysis and CA	20,000	413	1.0	1 MKOH	3500	—	—	[53]
Albizia flower	Porous carbon	One-step pyrolysis-and TA	10,000	406	0.5	1 M Na <sub>2</sub> SO <sub>4</sub>	2757.63	26.3	429	[54]
<i>Cucumis melo</i> fruit peel	AC	Carbonization and CA	8,000	404	1	1 M KOH	721.91	29.30	279.7	[55]
Celery	N-doped AC	Carbonization and CA under N <sub>2</sub> flow	10,000	402	1	1 M H <sub>2</sub> SO <sub>4</sub>	—	—	—	[56]
Spruce bark	Graphene nanosheet	HC and CA	10,000	398	0.5	6 MKOH	2385	—	—	[57]
Green bristle grass seeds	N, S and O-doped porous carbon	Carbonization-and chemical treatment	10,000	391	0.5	6 MKOH	—	—	—	[58]

Corncob	AC	Carbonization and CA	—	390	0.5	1 M EMIMBF <sub>4</sub>	800	25	174	[59]
Glucose and amino urea	N-doped porous carbons	HC and CA	10,000	390	0.5	—	2594	—	—	[60]
Cotton fabric and ZIF-8	NC/CT	Chemical treatment, carbonization and TA under N <sub>2</sub> flow	—	390	—	PVA-H <sub>2</sub> SO <sub>4</sub>	—	23.4	91.4	[61]
<i>Lentisusedo des</i>	HPC	HC and CA	10,000	389	0.2	1 M H <sub>2</sub> SO <sub>4</sub>	1144	45.69	—	[62]
Indian Cake Rusk	AC	Carbonization and CA	6,000	381.0	1.7	1 M H <sub>2</sub> SO <sub>4</sub>	1413.0	—	—	[63]
Paper towel	N-doped porous carbon	Carbonization, CA and acid etching	10,000	379.50	1	6 M KOH	1,123.40	—	—	[64]
Lotus leaf	Porous carbons	Carbonization and CA	—	379	1	6 M KOH	2488	9.2	491	[65]
<i>Cladophora glomerata</i> (raw biochar)	Porous carbon	Slow pyrolysis and CA	5,000	376.7	1	3 M KCl	—	42.4	—	[66]
Pomelo peel	Porous carbon	HC and CA	5,000	374	0.1	6 M KOH	—	20	—	[67]

Lignin derivative	N-doped HPC	Hydro thermal cross-linking reaction and CA	3,000	370	0.5	6 MKOH	1867.4	18.5	300	[68]
Waste coffee beans	Porous carbon	Chemical treatment and carbonization under N <sub>2</sub> flow	10,000	368	—	1 M H <sub>2</sub> SO <sub>4</sub>	—	20	—	[69]
Sawdust	N-doped AC	Polymerization and CA	10,000	367	0.5	6 MKOH	1674	8.7	251.6	[70]
Acetylene gas	Porous carbon	Chemical vapor deposition	—	362.6	1	—	920.4	24.4	8800	[71]
Cotton fabric	N-doped porous carbon	Carbonization in N <sub>2</sub> and TA	5,000	360	0.5	6 MKOH	777	—	—	[72]
Waste com silks	N-doped AC	Carbonization and CA	5,000	358	0.5	1 M H <sub>2</sub> SO <sub>4</sub>	1764.8	7.9	—	[73]
Prawn shell ( <i>Bohail prawn</i> )	N-doped AC	Simultaneous carbonization and CA	—	357	0.05	6 MKOH	—	—	—	[47]
<i>Arganiaspinosa</i> seed shells	N-rich AC	Carbonization and CA	—	355	0.125	1 M H <sub>2</sub> SO <sub>4</sub>	—	—	—	[74]
Waste Coca Cola	N,S dual doped AC	HC and TA	—	352.7	1	6 MKOH	—	—	—	[75]
<i>Cotonier spp.</i> fiber	AC	Carbonization and CA	10,000	346.1	1	6 MKOH	—	33.04	160	[76]

Water hyacinth	HPC	Carbonization and CA	10,000	344.9	0.5	—	2276	23.8	15700	[77]
Bio-oil	HPC	Template carbonization	—	344	0.5	6 MKOH	1409	8.1	638	[78]
Aniline modified lignin	Porous carbon	Carbonization and CA	1,000	336	1	6 MKOH	2265	—	—	[79]
Sucrose	N-doped porous carbon	Polymerization and CA	10,000	335	1	6 MKOH	1751	14.1	—	[80]
Polymeric resin	N-doped porous carbon	Carbonization and CA	—	333	1	6 MKOH	1946	—	—	[81]
BTA	N-doped porous carbon	Pyrolysis and CA	5,000	332	0.5	2 MKOH	1228	12.94	375	[82]
Soybean meal	N and O doped porous carbon	CA under-N <sub>2</sub> atmosphere	10,000	330	0.5	3 MKOH	1175	8.4	—	[83]
DTPA	N-doped porous carbon	Thermal treatment-and CA	5,000	323	0.5	6 MKOH	3214	—	—	[84]
Porcine bladders	N-doped porous carbon	Carbonization and CA	5,000	322.5	0.5	6 MKOH	1881.7	10.9	150	[85]

Seafood (crab)	AC	Carbonization, acid-etching and CA	20,000	320.4	1	6 MKOH	—	22.3	497	[86]
Bagasse wastes	BDHSC	HC and CA	15,000	320	0.5	1 M Na <sub>2</sub> SO <sub>4</sub>	2296	20	182	[87]
Chitosan	N and B co-doped AC	Carbonization, CA and hydrothermal doping	5,000	316	0.2	6 MKOH	1129.6	—	—	[88]
PMMA-PAN core-shell	HPC	Polymerization and carbonization under-N <sub>2</sub> atmosphere	2,000	314	0.5	6 MKOH	2085	—	—	[89]
Glucose-amine	N-doped AC	HC and CA under N <sub>2</sub> flow	—	313	0.5	6 MKOH	1836	7.2	250	[90]
Polymetric resin	N-doped porous carbon	Carbonization and CA	5,000	312	0.5	7 MKOH	1946	—	—	[81]
Glucose	N-doped AC	Chemical-blowing-and CA	4,000	312	0.5	6 MKOH	1997.5	—	—	[91]
Natural casings	N-enriched AC	Carbonization and CA	—	307.5	0.5	6 MKOH	3100	11.6	297	[92]
Waste dyed-silk fabrics	HPC	One-step pyrolysis	5,000	305.02	—	1 M Na <sub>2</sub> SO <sub>4</sub>	285.1	10.73	160.71	[93]
Porous starch	HPC	Carbonization and CA	10,000	304	0.05	6 MKOH	3251	—	—	[94]

<i>Euonymus japonicus</i> leaves	N and O doped micro-porous carbons	Pyrolysis and CA	5,000	303	0.2	6 MKOH	1268	15.2	60.3	[95]
Chitosan protic salt	N and S-doped HPC	Double-soft template solvent-free self-assembly method	5,000	302	—	6 MKOH	927	—	—	[96]
Waste bamboo	BHNC	Chemical treatment and CA	20,000	301	0.1	6 MKOH	1472	6.1	—	[97]
Sugarcane bagasse	N-doped porous carbon	Carbonization and CA	5,000	298	1	1 M H <sub>2</sub> SO <sub>4</sub>	—	27.7	—	[98]
Willow catkin	N and S doped PCNs	One-step pyrolysis and CA	10,000	298	0.5	6 MKOH	—	21.0	180	[99]
Petroleum pitch	PGCNs	Template carbonization and CA under Ar flow	7000	293	0.05	6 MKOH	1542-2305	—	—	[100]
Egg yolk	Porous carbon	Pre-carbonization, chemical treatment and carbonization under an argon atmosphere	2,000	287.66	0.5	6 MKOH	2277.79	—	—	[101]
Pigskin	N and O doped HPC	Carbonization and CA under N <sub>2</sub> flow	10,000	287.1	1	6 MKOH	—	43.0	875.0	[102]



Lignin	HPC	Carbonization, CA and post-chemical treatment	—	286.7	0.2	6 M KOH	3775	—	[103]
Tobacco rods	ACNS	HC and CA	10,000	286.6	0.5	6 MKOH	—	31.3	[104]
<i>Cetiba insignis</i> fiber	N-doped Porous carbon	Pre-treatment and one-step pyrolysis	2,000	283	1	6 MKOH	1125.7	—	[105]
<i>Moringaoleifera</i> stems	PCNSs	One-step pyrolysis and no-post-activation	20,000	283	0.5	1 M Na <sub>2</sub> SO <sub>4</sub>	2250	—	[106]
PC	AC	Carbonization and CA	—	277	0.5	6 MKOH	808	277	[107]
Waste sugar solution	Porous carbons	Two-step carbonization and CA	5,000	273.31	1.5	—	1953	5.09	[108]
Collagen waste	Mn-doped /N-containing carbon	Metal-ion chelation	6,000	272.62	1.0	1 MKOH	—	—	[109]
C <sub>2</sub> H <sub>4</sub>	CNTs/GNFs	Pyrolysis and carbonization	10,000	270	1	6 MKOH	1863.1	72.2	[37]
Polyethylenimine	N-doped AC	Centrifugation and carbonization under Ar	15,000	268	—	EMIMBF <sub>4</sub> /PC	—	60.31	[110]

Petroleum pitch	PGCMs	Template carbonization and CA under Ar flow	—	267	0.05	BMIMPF <sub>6</sub>	1542-2305	148.3	204.2	[100]
Coconut fiber	AC	Carbonization and CA	—	266	0.1	6 M KOH	2898	53	8224	[111]
Aloe peel	Porous carbons	HC and CA	5,000	264	0.5	—	1286	—	—	[112]
PAN/DMF	G/CNFs	Electro-spinning and carbonization	2,000	263.7	0.1	—	—	—	—	[113]
Cigarette filter	N-doped HPC	Carbonization and CA	5,000	263	1	—	3420	—	—	[114]
Wastes corn silks	N-doped AC	Carbonization and CA	1,000	260.8	0.5	Alizarin-red, bromoamine acid in H <sub>2</sub> SO <sub>4</sub>	1764.8	17.8	—	[73]
Bio-tar	Porous carbon	Carbonization and CA	5,000	260	0.25	1 M TEABF <sub>4</sub> /AN	2546	17.8	—	[115]
polymer nanotubes	S-doped CNTs	Carbonization and CA	1,000	259	1	6 M KOH	—	—	—	[116]
<i>Arganiaspinosa</i> seed shell	O-rich AC	Carbonization and CA	—	259	0.125	1 M H <sub>2</sub> SO <sub>4</sub>	2100	—	—	[74]
Wax gourd	Porous carbon	Carbonization and CA	—	258	1	KOH/PVA	2919	13.0	297.5	[117]

Pinecone	AC	Carbonization and CA	20,000	255	1.4	PVDF-HFP/EMP/TFSL/ SN/LiTFSI	1781	20	55700	[118]
Walnut shells	Porous carbon	HC and CA under N <sub>2</sub> flow	3,000	255	0.5	—	—	—	—	[119]
Cotton stalk	FTMAC	Carbonization and CA	10,000	254	0.2	1 M H <sub>2</sub> SO <sub>4</sub>	1964.46	18.14	450.37	[120]
Pinecone	AC	Carbonization and CA	20,000	244	1.4	PVDF-HFP/EMP.TF/S N	1781	19	39300	[118]
Lotus pollens	ACMs	Pre-treatment, carbonization and CA	10,000	244	0.1	1 M H <sub>2</sub> SO <sub>4</sub>	—	—	—	[121]
Coal tar pitch	IMCSs	Pulverization and CA under N <sub>2</sub> flow	10,000	242	0.05	6 MKOH	1 046	—	—	[122]
Pomelo peels	N-and dual-doped AC	Chemical treatment and carbonization	10,000	240	0.5	2 MKOH	807.7	11.7	160	[123]
Mango stern peel	Porous carbon	HC and pyrolysis	—	240	1	6 MKOH	1270	—	—	[124]
Spruce bark	Graphene nanosheet	HC and CA	—	239	1	1M TEABF <sub>4</sub> /AN	2385	74.4	743.7	[57]
Waste shrimp shells	N, and O-doped porous carbon	CA and carbonization	5,000	239	0.5	6 MKOH	1271	—	—	[125]

Glucose	P, N dual-doped holey graphene	Chemical-treatment-and carbonization	25,000	235.5	0.5	1 M TEABF <sub>4</sub>	—	—	[126]
Sucrose	N-doped HPC	Pyrolysis and CA	20,000	229	0.1	6 MKOH	2160	—	[127]
ZIF-7	Porous carbon	Carbonization of-ZIF-7 and additional carbon sources	5,000	228	0.1	6 MKOH	—	—	[128]
Natural almond	N-doped porous carbon	In-situ-hard template and CA	—	228	1	6 MKOH	1877.8	—	[129]
Resole	HPCs	CA and thermal treatment under nitrogen protection	5,000	226	0.05	7 MKOH	1201	—	[130]
Puffed rice	N-doped CNS	Carbonization and CA	—	218	80	6 MKOH	3326	104	[131]
Indian Cake Rusk	AC	Carbonization and CA	10,000	217.0	0.35	1M LiPF <sub>6</sub>	1413.0	47.1	[63]
GO paper	r-GO	Flame-induced reduction of GO-paper	2,000	212	1	2 MKOH	274.9	—	[132]
Furfural residues	Porous carbon	Carbonization and CA	10,000	210.2	—	6 MKOH	2494	52.6	[133]

<i>Enteromorpha prolifera</i>	HPCs	Carbonization and CA under nitrogen	—	210	3	6 MKOH	3332	—	[134]
Urea and humic acid	N-doped HPC	Direct CA under N <sub>2</sub> flow	10,000	209	0.05	6 MKOH	3142	88	[135]
Squidgladius chitin	O and N enriched AC	Carbonization-and pyrolysis under nitrogen atmosphere	25,000	204	—	1 MH <sub>2</sub> SO <sub>4</sub>	1129	4.53	9900 [136]
CBAP	N-doped CBAP	Chemical treatment and carbonization	—	203.2	1	6 MKOH	1060	—	[137]
Gelatin	N-doped meso-porous carbons	Carbonization-under N <sub>2</sub> flow	5,000	198	20	6 MKOH	666	—	[138]
<i>Cladophora glomerata</i> (treated biochar)	Porous carbon	Slow pyrolysis and CA	—	197.8	1	3 M KCl	—	22.25	450 [66]
Bamboo	AC	Carbonization and CA under N <sub>2</sub> flow	10,000	193.8	20	3 MKOH	2221.1	10.9	63 [139]
PVA/GO	Graphene-doped porous carbon	HC and CA	—	190	1	GO-doped-ion gel	3193	76	840 [140]
Banana	N-doped porous carbon	Carbonization and CA	—	178.9	1	6 MKOH	1414.97	—	— [141]

<i>Pongamia pinnata</i> seed oil	HPC	Carbonization and CA	3,000	177	—	1M KOH	1065	44	18000	[142]
Xinjiang coal	B-N co-doped AC	Carbonization and TA under argon flow	20,000	176	0.5	6 M KOH	—	9.7	2000	[143]
Industrial GO powder	N-doped graphene aero gels	Solid support method	5,000	175	0.5	6 M KOH	—	—	—	[144]
Lignin	O-enriched porous carbon	Carbonization and CA	—	173	0.5	PVA/LiCl	—	23.0	—	[145]
Corn syrup	AC	HC with TA under N <sub>2</sub> flow	—	168	0.2	6 M KOH	1473	4.2	1500	[146]
Carton box	AC	Carbonization and CA under nitrogen atmosphere	—	168	0.5	1 M TEABF <sub>4</sub> /AN	2731	45	338	[147]
Peanut shell	AC	Ethanol soaking with HC and CA	—	168	—	PVDF-HFP/EMTFMg (Tf) <sub>2</sub>	1483	—	—	[148]
Wax gourd	Porous carbon	Carbonization and CA	—	167	1	0.5 M Na <sub>2</sub> SO <sub>4</sub>	2919	16.3	8625	[117]
Hemp stem	AC	HC and CA	—	167	1	1.8 M TEMABF <sub>4</sub> /PC	2801	19.8	21000	[149]
Cellulose	MMC	Carbonization without further activation	—	160	0.2	—	646	17.81	180.11	[150]

GO paper	r-GO	Flame induced reduction of GO-paper	—	160	1	1 M Et <sub>4</sub> NBF <sub>4</sub>	—	—	[132]
Green onion leaves	Porous carbon	Carbonization and TA	—	158.6	0.2	3 MKOH	551.7	—	[151]
CBAP	N,S-doped CBAP	Chemical treatment, and carbonization	—	157.7	1	6 MKOH	1047	—	[137]
Mango kernel	AC	Carbonization, pulverization and CA	5,000	157.5	—	1 M H <sub>2</sub> SO <sub>4</sub>	—	11.8	[29]
Coconut fiber	HPAC	Carbonization and CA	—	155	0.1	1 M EMIMBF <sub>4</sub>	2898	—	[111]
Cigarette filter	N-doped porous carbon	Carbonization under Ar atmosphere	6,000	153.8	1	6 MKOH	1634	—	[152]
Fibers of empty palm fruit bunch	AC	Carbonization and CA	—	150	—	1 M H <sub>2</sub> SO <sub>4</sub>	1704	4,297	[153]
Tobacco	AC	Carbonization and CA under-N <sub>2</sub> atmosphere	9,000	148	0.5	6 MKOH	1297.6	—	[154]
Peanut-shell	AC	Ethanol soaking with HC and CA	—	148	—	PVDF-HFP/EMITf	1411	—	[148]
Rice husk	AC	Carbonization and CA	10,000	147.0	0.1	6 MKOH	2696	5.11	[155]

Corn starch	Porous carbons	HC and CA	—	144	0.625	6 MKOH	1239	19.9	311	[156]
Waste tyres	AC	Carbonization and CA under N <sub>2</sub> flow	1,000	140	0.25	1 M K <sub>3</sub> Fe(CN) <sub>6</sub> -1 M HNO <sub>3</sub>	147-385	—	—	[157]
Polystyrene-e	Porous carbon sheets	Template Carbonization - and chemical activation	10,000	135	1	1 MH <sub>2</sub> SO <sub>4</sub>	—	3.4	250	[158]
Rubber wood waste	Porous carbon	Chemical treatment and carbonization under N <sub>2</sub> flow	—	129	—	1 MH <sub>2</sub> SO <sub>4</sub>	—	14.2	500	[159]
Hemp	CNS	Carbonization and CA	10,000	122	1	BMPY.TFSI	280	34	20000	[160]
Waste cotton	AC	Carbonization and CA	10,000	112	1	1 M TEABF <sub>4</sub> /AN	1550	29.50	—	[161]
Polyphosph-azene	N, P and O- nonporous carbon	Direct carbonization and CA	10,000	105	0.3	6 MKOH	1798	—	—	[162]
<i>Saccharumb engale-nse</i>	AC	CA	120,000	102.6	—	1 MLi <sub>2</sub> SO <sub>4</sub>	2090	—	—	[25]
Lofa sponge	AC	Carbonization and CA	—	93	0.1	10 mM NaCl	1819	—	—	[163]



p-nitrobenzena mine	N,S doped nonporous carbon	Carbonization	—	73	1	6 MKOH	—	—	[164]
Tremella	AC	Carbonization and CA	10,000	71	1	6 MKOH	3760	65.6	19,700 [165]
Tremella	AC	Carbonization and CA	—	60	0.5	1 M Na <sub>2</sub> SO <sub>4</sub>	3760	28	— [165]
Baobab Fruit Shells	AC	Carbonization and CA under argon flow	1000	58.67	1	6 MKOH	—	20.86	400.09 [166]
Waste compact discs	AC	Thermal-activation under nitrogen atmosphere	—	51	—	—	1214.25	21.43	700 [167]
Kapok fibers	Meso-porous carbon flakes	Carbonization and CA	5,000	—	—	1 M TEABF <sub>4</sub> /PC	3,010	33.8	260 [168]
Polystyrene foam	N-doped AC/G	Chemical treatment, carbonization and CA	—	—	0.05	6 MKOH	—	11.8	12.5 [169]
Chitosan protic salt	N and S-doped HPC	Double-soft template solvent free self-assembly method	—	—	—	1 M Na <sub>2</sub> SO <sub>4</sub>	927	17.6	250 [96]
Waste bamboo	BHNC	Chemical treatment-and CA	—	—	—	6 MKOH	1472	43.3	— [97]

Oil-palm kernel shell	AC	Pyrolysis and CA	—	—	—	1 M Na <sub>2</sub> SO <sub>4</sub>	—	6.2	300	[170]
Palm kernel shell	AC	Pyrolysis and CA	—	—	—	1 M Na <sub>2</sub> SO <sub>4</sub>	—	7.4	300	[170]
Lotus leaf	Porous carbons	Carbonization and CA	—	—	—	1 M Na <sub>2</sub> SO <sub>4</sub>	2488	23	868	[65]
Mustard oil	Carbon aerogel	Thermal treatment and CA	—	—	—	H <sub>2</sub> SO <sub>4</sub> -PVA	1032	55.6	7900	[171]
MRF	NGHPC	Carbonization, graphitization and CA	10,000	—	—	0.5 M Na <sub>2</sub> SO <sub>4</sub>	2729	33.9	809.9	[52]
Silkworm cocoon	N-doped porous carbon	Pre-carbonization, carbonization and CA	—	—	—	1 M TEABF <sub>4</sub> /AN	3386	34.41	31250	[172]
Silkworm cocoon	N-doped porous carbon	Pre-carbonization, carbonization and CA	—	—	—	EMIMBF <sub>4</sub>	3386	112.1	23910	[172]
Glucose	N-doped AC	Chemical blowing-and CA	—	—	—	0.5 M Na <sub>2</sub> SO <sub>4</sub>	1997.5	20.2	448	[91]
Green bristle grass seeds	N, S and O-doped porous carbon	Carbonization and CA	10,000	—	—	1 M Na <sub>2</sub> SO <sub>4</sub>	—	20.15	500	[58]
Starch	N-doped PCNs	Chemical blowing, graphitization and CA	5000	—	—	1 M TEABF <sub>4</sub> /AN	—	27.5	—	[173]

Carbon nanosheet (CNS), carbon nanotubes (CNTs) bagasse-derived hierarchical structured carbon (BDHSC), activated carbon (AC), AC/graphene (AC/G), hierarchical porous carbon (HPC), carbon microsphere/polyaniline (YC/PANI), diethylenetriaminepentaacetic acid (DTPA), melamine resorcinol formaldehyde (MRF), bio-inspired beehive like hierarchical nanoporous carbon (BHNC), flute type micropores activated carbon (FTMAC), porous carbon nanosheets (PCNs), carbonyl-based aromatic porous polymer (CBAP), poly(vinyl alcohol) (PVA), nitrogen-doped porous carbon nanotubes/carbon fabric (NC/CT), zeolitic imidazolate framework (ZIF-8), activated carbon microspheres (ACMs), interconnected mesoporous carbon sheets (IMCSs), micro- and meso-porous carbon (MMC), hierarchically porous carbon nanosheets (PCNSs), nitrogen-enriched graphitized hierarchical porous carbon (NGHPC), activated carbon fibers (ACFs), pinecone biomass (PC), porous graphene-like carbon materials (PGCMs), poly(methylmethacrylate) (PMMA), polyacrylonitrile (PAN), graphene oxide (GO), reduced graphene oxide (r-GO), activated carbon nanospheres (ACNS), graphene beaded carbon nanofibers (G/CNFs), N,N-dimethylformamide (DMF), 1H-benzotriazole (BTA), graphitic nanofibers and carbon nanotubes (GNFs/CNTs), hydrothermal carbonization (HC), chemical activation (CA), thermal activation (TA), 1-butyl-3-methylimidazolium hexafluorophosphate (BMIMPF<sub>6</sub>), poly(vinylidene fluoride-co-hexafluoropropylene) (PVDF-HFP), lithium bis(trifluoromethanesulfonyl) imide (LiTFSI), trifluoromethanesulfonyl imide (EMP.TFSI), 1-ethyl-3-methylimidazolium trifluoromethanesulfonate (EMITf), magnesium trifluoromethanesulfonate (Mg (Tf)<sub>2</sub>), 1-butyl-1-methylpyrrolidinium bis(trifluoromethylsulfonyl) imide (BMPY.TFSI), 1-Ethyl-3-methylimidazolium tetrafluoroborate in propylene carbonate (EMIM BF<sub>4</sub>/PC).

### Conclusion and recommendations

It has been shown that research on energy conversion and storage, particularly using SCs, is growing progressively. A concise description of different capacitor types was given, with a focus on the non-Faradaic SCs. Review of recent carbon electrode materials with their practical performances (specific surface area, specific capacitance, energy and power densities) for non-Faradaic SCs, was made.

Development and investigation of highly active carbon materials with optimized electrolytic compatibilities and manipulative morphologies and pore structures is recommendable. In this regard, certain materials of renowned potentials are yet to be fully engaged as electrode materials for SCs and they include hemp (*Cannabis sativa*) and cigarette filters. A team of Canadian researchers utilized hemp fibers via hydrothermal carbonization (HC) combined with activation and found the material as efficient as graphene, in SC application [30, 160, 174]. Just afterward, "Used cigarette filters turned into supercapacitor electrodes that outperform graphene-based ones" [175], as Korean researchers utilized carbon materials prepared by a novel one-step method from cigarette filters and used it in SC application. It exhibited higher capacitance than N-doped graphene or N-doped carbon nanotube electrodes [30, 175, 176]. These materials and the likes should be explored.

Finally, in a very recent study by Jain *et al.* [29], activated carbon electrode was used to achieve both EDL super-capacitance and hybrid super-capacitance, by simply adding a redox agent (p-hydroxyaniline) to the electrolyte (aqueous sulphuric acid), rather than employing the synergistic effect of different electrode materials [4,8,177]. This can pave the way for new designs of hybrid SCs with unique features and therefore, further investigation is recommended.

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