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Carbonaceous electrode materials for supercapacitor: Preparation and surface functionalization

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Supercapacitors became more and more important recently in the area of energy storage and conversion. Their large power deliveries abilities, high stability and environmental friendliness characteristics draw tremendous attention in high-power applications such as public transit networks. Carbonaceous materials with unique surface and electrochemical properties were widely used in supercapacitors as electrode materials. This review focuses on the developments in supercapacitor electrodes made from carbonaceous materials recently, their working principle and evaluation parameters were summarized briefly. The preparation methods and electrochemical properties of different carbonaceous materials were compared and classified. It was found that the surface situation (e.g., porous structure, hydrophilic) of carbonaceous materials strongly affect the electrochemical performances of supercapacitor. So far, active carbons would be the most applicable carbonaceous electrode materials owing to their good chemical stability and conductivity, extensive accessibility inexpensiveness. But their energy densities still fall behind practical demands. Both theoretical calculations and experimental studies show that surface modification and doping of carbonaceous materials can not only optimize their pore size, structure, conductivity and surface properties, but also can introduce extra pseudocapacitance into these materials. Considering global environmental pollution and energy shortage problems nowadays, we sincerely suggested that future work should focus on domestic, medical and industrial wastes residues derived carbonaceous materials and scaled production process such as reactors and exhaust gas treatment.

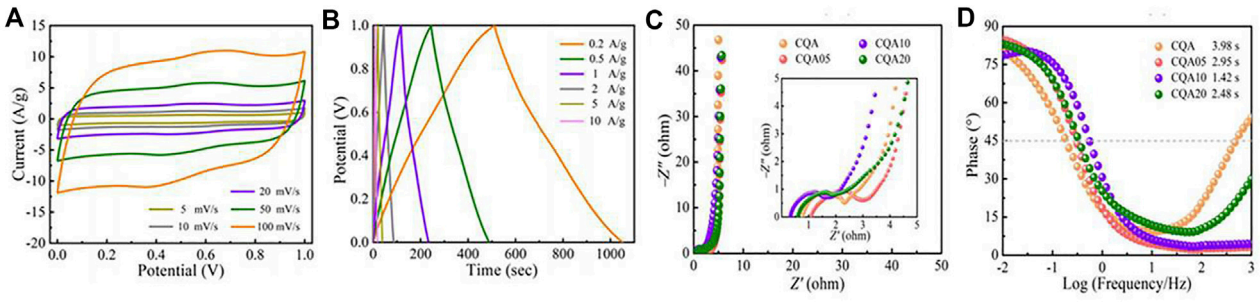
KEYWORDS

supercapacitors, electrode materials, surface functionalization, working principal, EDLC

1 Introduction

Over the past two decade, the comsumption and cost of gradual exhausted fossil fuels were significantly increased due to industrial developments and international competitions (Sujatha, 2021). Moreover, abuse of fossil fuels have being caused environmental pollution and global warming (Welsby et al., 2021). In order to cope these crises, wind energy, solar energy and other unstable but sustainable energy sources are urgently needed (Zhang et al., 2017; Chen et al., 2020; Shao et al., 2020). Therefore, efficient installation for energy storage, including and supercapacitors (or electrochemical capacitors) and batteries, have attracted tremendous attentions. Supercapacitors have unique advantages especially in terms of wide operating temperature range, no memory effect, good cycling stability and high watt density (Gao et al., 2013; Lee et al., 2017; Song et al., 2022a). The electrochemical performance comparison about supercapacitors, conventional capacitors and batteries were listed in Supplementary Table S1 (Salunkhe et al., 2016). Overall performance of supercapacitors currently lies between traditional dielectric capacitors and batteries. Generally the energy density of supercapacitors is relatively lower than that of many batteries, but the unique electricity storage process enables supercapacitors to store or release substantial amount of electricity in a very short time, which let supercapacitors become a most competitive candidate for the technologies that need are fast to transport and require a quick surge of electrical energy in a very short moment such as wind turbines, grid stabilization system, uninterruptable power supply, forklifts, military weapons, load cranes, hybrid electric vehicles, regenerative braking systems for electric vehicles and rail transit (Taberna et al., 2003; Meng et al., 2004; Chmiola et al., 2010; Wei et al., 2010; Cheng et al., 2011; Lv et al., 2020; Hasan Md et al., 2022).

So far, the working principle of common supercapacitors and electrode materials have been fully studied. Electrical energy can be stored in the form of capacitance by supercapacitors *via* storing positive and negative charges on their spaced opposing electrodes. Supercapacitors falls into two categories based on different capacitance storage principles: "Electric double layer capacitors (EDLC)" and "Faraday quasi capacitors" (or named pseudocapacitors) (Luryi, 1988; Pekala, 1989; Conway et al., 1997; Frackowiak, 2007; Chmiola et al., 2010; Stoller et al., 2011; Paek et al., 2013; Liang, 2015; Zhu et al., 2016; Chen et al., 2019; Ghosh et al., 2019; Xue et al., 2019; Ghosh et al., 2020; Miao et al., 2021a; Song et al., 2021a; Wang et al., 2021a; Miao et al., 2021b; Song et al., 2021b; Fu et al., 2021; Long et al., 2021; Mansuer et al., 2021; Sakita et al., 2021; Xin et al., 2021; Zhang et al., 2021; Yang et al., 2022a; Du et al., 2022; Mansuera et al., 2022; Qin et al., 2022; Yan et al., 2022; Zheng et al., 2022; Maldonado-Hódar et al., 2000; Haddon, 2002; Lachawiec et al., 2005; Ji et al., 2009; Qin et al., 2009; Li et al., 2011; Lv et al., 2012; Morteza et al., 2015; Mousavi-Khoshdel and Targholi, 2015; Mousavi-Khoshdel et al., 2016; Ping et al., 2016; Wang and Pumera, 2016; Liu et al., 2017; Song et al., 2018; Silva et al., 2020; Kolosov and Glukhova, 2021a; Kolosov and Glukhova, 2021b; Boulanger et al., 2021; Cao et al., 2021; Chakrabarty et al., 2021; Chen et al., 2021; Christians et al., 2021; Song et al., 2021c; Hoa et al., 2021; Nizam et al., 2021; Olabi et al., 2021; Ramirez et al., 2021; Tong et al., 2021; Vermisoglou et al., 2021; Zhai et al., 2021; Zhou et al., 2021; Yang et al., 2022b; Mei, 2022; Rashidi et al., 2022; Teimuri-Mofrad et al., 2022; Liu et al., 2022; Paul1 and Roy, 2021; Basivi et al., 2021; Zhang et al., 2019; Zhai et al., 2011; Rashidi and Yusup, 2020; Yang and Zhou, 2017; Arango et al., 2018; Umezawa et al., 2021; Mandal et al., 2021; Wulandari et al., 2021; Wu et al., 2021; Taer et al., 2021; Kalu-Uka et al., 2022; Yuan et al., 2021; Gopalakrishnan and Badhulika, 2020; Wang et al., 2015; Du et al., 2021; Jin et al., 2019; Hassan et al., 2014; Kakaei et al., 2016; Kumari et al., 2018; Hummers and Offeman, 1958; Becerril et al., 2008; Wang et al., 2009; Kaskela et al., 2010; Liu et al., 2011; Sun et al., 2013; Gao et al., 2014; Long et al., 2015; Baig et al., 2016; Xiao et al., 2016; Li et al., 2017; Tian et al., 2017; Wang et al., 2017; Kaur et al., 2018; Sahoo et al., 2018; Hoang et al., 2019; Tao et al., 2019; Wang et al., 2019; Du et al., 2020; Kakaei et al., 2020; Wang et al., 2021b; Pang et al., 2021; Singh et al., 2021; Tatrari et al., 2021; Zhao et al., 2021; Akin and Zhou, 2022; Balboni et al., 2022; Song et al., 2022b; Elanthamilan et al., 2022; Zhai et al., 2022). As shown in Figure 1, to evaluate the performance of supercapacitors, several Electrochemical analysis methods such as cyclic voltammetry (CV), electrochemical impedance spectroscopy (EIS) and galvanostatic charge-discharge (GCD) test and were widely used. It has been found that important parameters of supercapacitors (e.g., operating potential window, equivalent series resistance and capacitance), are strongly depends on their electrode materials (Frackowiak, 2007; Ghosh et al., 2019). As shown in Figure 1A, capacitance (C) of supercapacitors could be obtained from CV curves using Eq. 1, where ΔV is the total potential bias of the voltage window, dt is the sampled span of time, i is a sampling current. If CV curves are close to ideal rectangles, C could be calculated using Eq. 2, where ν is the potential scan rate. Gravimetric capacitance (C_m) is an important parameter for supercapacitors, when CV curves of a supercapacitor show the characteristics of a non-ideal rectangle (e.g., have pseudocapacitive bulging peaks), C_m of the electrode materials could be calculated from enclosed area between the current and the voltage in the CV curve using Eq. 3, where m is electroactive materials' mass in electrodes, $I(V)$ denotes the density of response current, V_c and V_a are CV curve's integration limits (Wei et al., 2010). As shown in Figure 1B, the GCD curve can calculate the C_m of the electrode material (4), where Δt *is* the time of discharge, m is the active materials' mass, ΔV is the change of voltage which is excluded voltage drop (IR drop) in the process of discharge, I is the current of discharge. After assembling into a symmetrical supercapacitor, the capacitance of single electrode (C_s) can be calculated through



$$C = \Delta Q / \Delta V = \left(\int i dt\right) / \Delta V \quad (1) \qquad C = \frac{\Delta Q}{\Delta V} = i\frac{\Delta t}{\Delta V} = i / \nu \quad (2) \qquad C_m = \frac{1}{m\upsilon\,(V_c - V_a)} \int_{V_a}^{V_c} I(V)\,dV \quad (3)$$

$$C_m = \frac{I \times \Delta t}{m \times \Delta V} \quad (4) \qquad C_s = \frac{4 \times I \times \Delta t}{m \times \Delta V} \quad (5) \qquad C'(\omega) = \frac{-Z''(\omega)}{\omega |Z(\omega)|^2} \quad C''(\omega) = \frac{-Z'(\omega)}{\omega |Z(\omega)|^2} \quad (6)$$

$$C_m = \frac{2 \cdot |\ Z''\ |}{2\pi f \cdot \left[(\ Z''\ )^2 + (\ Z'\ )^2\right] m} \quad (7) \qquad E = \frac{1}{2} C \Delta V^2 \quad (8) \qquad P = \frac{E}{\Delta t_d} \quad (9) \qquad \eta = \frac{\Delta t_d}{\Delta t_c} \quad (10)$$

FIGURE 1
An expression of a typical curves of **(A)** CV and **(B)** GCD, **(C)** Nyquist plots, **(D)** Bode phase diagram (Mansuera et al., 2022); equations for calculations of capacitances (C, C_m, Cs, C′, C″), energy and power density (E, P) as well as coulombic efficiency (η) (Taberna et al., 2003; Wei et al., 2010; Cheng et al., 2011).

Eq. 5, where m represents the gross mass of active materials on two working electrodes, but I, t, and V reflect same meaning as Eq. 4. Volumetric capacitance (C_V) and areal-specific capacitance (C_A), which can be obtained according to the volume or the area of the single electrode through Supplementary Eqs S1, S2, are also important. Since the thickness of electrode materials are difficult to be measured accurately till now, compare to C_m much fewer studies reported C_V or C_A (Li et al., 2017; Zhao et al., 2021). EIS provides focused analysis of the resistive behavior. Solution resistance, charge transfer impedance, could be calculated from Nyquist plots' altofrequency part (Figure 1C), while low frequency part of the curve shows the ion diffusion impedance. In addition, the Bode phase diagrams (Figure 1D) show phases angle as a function of frequency. The capacitance's real and imaginary parts ($C'_{(\omega)}$ and $C''_{(\omega)}$) can be calculated from the impedance data based on Eq. 6, where complex impedance is $Z_{(\omega)}$, its real and imaginary parts are $Z'_{(\omega)}$ and $Z''_{(\omega)}$, and the angular frequency is ω (Taberna et al., 2003). There is a Eq. 7 can also be used to calculate C_m, where m is active materials' mass in each electrode, f is the operating frequency, and Z'' and Z' are the imaginary and real parts of total resistance (Cheng et al., 2011). Besides, supercapacitors' energy density (E), power density (P) and coulombic efficiency (η) can be estimated by Eqs 8–10, where ΔV, Δt_d, and Δt_c denote the voltage window, discharge time and charge time (Mansuera et al., 2022).

Electrode material is a crucial component in supercapacitors. With fascinating characteristics of environmental benignity and abundance, carbonaceous materials equipped with extensive applicability and outstanding physicochemical stability have been one kind of the most popular materials that applied in supercapacitors nowadays. Since a satisfactory capacitance can be generated due to the high ionic accessible surface area of the electrode material in a certain electrolyte, carbonaceous materials with the large surface area have been extensively explored over the past two decades (Chen et al., 2020; Shao et al., 2020). Different dimensional (0D-3D) carbonaceous materials, such as zero-dimensional (0D) carbon quantum dots (CQDs) and carbon nanospheres, one-dimensional (1D) carbon nanotube (CNTs) and nanowires, two-dimensional (2D) graphenes and carbon nanosheets, three-dimensional (3D) carbide-derived carbons (CDCs), activated carbons (ACs) and carbon aerogels, have been developed and tested in supercapacitors' electrode (Luryi, 1988; Pekala, 1989; Conway et al., 1997; Stoller et al., 2011; Paek et al., 2013; Zhu et al., 2016; Chen et al., 2019; Xue et al., 2019; Ghosh et al., 2020; Miao et al., 2021a; Song et al., 2021a; Wang et al., 2021a; Miao et al., 2021b; Song et al., 2021b; Fu et al., 2021; Long et al., 2021; Mansuer et al., 2021; Sakita et al., 2021; Xin et al., 2021; Zhang et al., 2021; Yang et al., 2022a; Du et al., 2022; Mansuera et al., 2022; Qin et al., 2022; Yan et al., 2022; Zheng et al., 2022).

The main energy storage mechanism of supercapacitors prepared from 3D carbonaceous materials such as ACs and carbon aerogels is EDLCs (Fu et al., 2021). However, it has been found that low-demensional materials (e.g., 1D CNTs and 2D graphene) themselves' properties can also contribute capacitance to the supercapacitor system. Thus, quantum capacitance (C_Q) has been formally proposed (Luryi, 1988). C_Q originates from electronic density of states (DOS) which close to the Fermi level of electrode materials. Doping, functionalization and mechanical deformation can be used to adjust materials' electronic structure and C_Q (Stoller et al., 2011). As shown in Supplementary Figure S1, Paek et al. (2013) evaluated interfacial capacitance between graphene and [BMIM][PF_6] as a function of applied potential (Φ_a). Total capacitors (C_T) at the interface of graphene/[BMIM][PF_6] is give as a range of electric double layer capacitance (C_D) and the quantum capacitors (C_Q) of graphene. When the DOS of the electrode material is relatively small, the effect of C_Q can be ignored.

Nevertheless, insufficient energy density still have being an important defect for carbonaceous materials, which shortens the long-term availability of the energy provided by supercapacitors and fall behind practical demands (e.g., in electric vehicles and rail transit) (Chen et al., 2019). Previous studies have shown that the carbonaceous materials' performance can be improved *via* surface functionalization, and it was found that surface-active groups of carbonaceous supercapacitor materials is mechanically important, several recently reports had started to study the surface situations of carbon materials *via* quantum-chemical computing (Zhu et al., 2016). Recently, considerable progresses have been made in heteroatom doped and/or surface functionalized carbonaceous materials which have potential for improved ions accessibility, material conductivity, faradaic feature, supercapacitive activities as well as energy storage abilities (Ghosh et al., 2020).

Several reviews related with carbonaceous electrode materials have been reported already (Frackowiak, 2007; Wang et al., 2021b; Akin and Zhou, 2022; Song et al., 2022b; Zhai et al., 2022), which gave almost the complete picture of the technologies for supercapacitor. However, there are few reviews that comprehensively introduce the surface functionalization of electrode materials. This mini-review aims to focus recent developments in the supercapacitor electrodes made from carbonaceous materials. The preparation, surface functionalization as well as electrochemical performances of different carbonaceous electrode materials were compared and classified. The challenges and potential prospects for future innovations of carbonaceous electrode materials were also discussed.

2 Preparation and performance of carbonaceous electrode materials

Many sources of carbonaceous materials. Various natural resources, such as biomass materials, crude oil, nature gas and coal, artificial polymer such as polyvinylidene chloride (PVDC), can be used as precursors for carbonaceous materials. There are various methodologies for converting carbonic precursors into carbonaceous materials, such as physical or chemical vapour deposition, pyrolytic heating as well as organic redox methodologies (Chmiola et al., 2010; Pekala, 1989; Conway et al., 1997; Maldonado-Hódar et al., 2000; Haddon, 2002; Lachawiec et al., 2005; Ji et al., 2009; Qin et al., 2009; Li et al., 2011; Lv et al., 2012; Liang, 2015; Morteza et al., 2015; Mousavi-Khoshdel and Targholi, 2015; Mousavi-Khoshdel et al., 2016; Ping et al., 2016; Wang and Pumera, 2016; Liu et al., 2017; Song et al., 2018; Silva et al., 2020; Kolosov and Glukhova, 2021a; Boulanger et al., 2021; Cao et al., 2021; Chen et al., 2021; Christians et al., 2021; Hoa et al., 2021; Nizam et al., 2021; Olabi et al., 2021; Ramirez et al., 2021; Zhai et al., 2021; Zhou et al., 2021; Yang et al., 2022b; Mei, 2022; Rashidi et al., 2022; Teimuri-Mofrad et al., 2022; Kolosov and Glukhova, 2021b; Song et al., 2021c; Vermisoglou et al., 2021; Tong et al., 2021; Chakrabarty et al., 2021; Liu et al., 2022; Paul1 and Roy, 2021; Basivi et al., 2021; Zhang et al., 2019; Zhai et al., 2011; Rashidi and Yusup, 2020; Yang and Zhou, 2017; Arango et al., 2018; Umezawa et al., 2021; Mandal et al., 2021; Wulandari et al., 2021; Wu et al., 2021; Taer et al., 2021; Kalu-Uka et al., 2022; Yuan et al., 2021; Gopalakrishnan and Badhulika, 2020; Wang et al., 2015; Du et al., 2021; Jin et al., 2019; Hassan et al., 2014; Kakaei et al., 2016; Kumari et al., 2018; Kaur et al., 2018; Du et al., 2020; Hoang et al., 2019; Sahoo et al., 2018; Kakaei et al., 2020; Gao et al., 2014; Baig et al., 2016; Kaskela et al., 2010; Liu et al., 2011; Balboni et al., 2022; Tao et al., 2019; Singh et al., 2021; Hummers and Offeman, 1958; Becerril et al., 2008; Wang et al., 2009; Long et al., 2015; Pang et al., 2021; Wang et al., 2017; Sun et al., 2013; Tian et al., 2017; Wang et al., 2019; Elanthamilan et al., 2022; Xiao et al., 2016; Tatrari et al., 2021). Through controlling the carbonization process and conditions, carbonaceous materials with different electrochemical performance were prepared. For instance, by altering the heating temperature, pressure and atmosphere in pyrolysis process, the properties of resulted carbonaceous materials can be adjusted. Carbonization process of carbonaceous materials is usually accompanied with graphitization which realize the orderly transformation of carbon atoms from a thermodynamically unstable turbostratic framework to a graphite structure. The degree of graphitization alters the performance of electrode material including comprehensive resistance, power delivery, energy efficiency, power density, cyclic stability and rate capacity. As the graphitization degree increased, carbonaceous materials' conductivity will be improved, but their relative porous structure may get distorted along with decreased specific area (Zhang et al., 2017).

2.1 CQDs and other 0D carbonaceous materials

CQDs are a typical class of 0D carbonaceous materials, which are composed of dispersed, quasi-spherical and ultrafine carbon nanoparticles with an average size below 10 nm (Hassan et al., 2014). CQDs can be obtained through different carbon sources (e.g., graphene feathers, egg, garlic, citric acid, etc.) *via* different methods (Kakaei et al., 2016; Kaur et al., 2018; Kumari et al., 2018; Du et al., 2020).

Application of pure CQDs as electrode materials for supercapacitors has not received much attention. But more and more CQDs based composites show excellent electrochemical performance for supercapacitors. Because of p–p conjugation, functional groups and strong electrostatic interaction of CQDs, a number of molecules such as organic molecules, nanoparticles, and polymers are able to connect to the surface of CQDs. For example, Hoang et al. (2019) combined CQDs and reduced graphene oxide (RGO) with a 1:2 mass ratio, this CQDs/RGO composite revealed high specific capacities (maximum capacitance of 278 F/g at the current density of 0.2 A/g) as electrode material for supercapacitors. As shown in Supplementary Figure S2, Sahoo et al. (2018) prepared a nanostructured CQDs/NiS composite through a facile hydrothermal method. This composite material exhibited good capacitive performance with specific capacity of 880 F/g at the current density of 2 A/g (Sahoo et al., 2018). Kakaei et al. (2020) presented a CQDs/PANI composite using CQDs as the shell and PANI as the core. This composite material showed good electrochemical performance with specific capacitance of 264.6 F/g at the current density of 2.5 A/g (Kakaei et al., 2020).

The diameter of other 0D carbonaceous materials usually larger than 50 nm. Compare with solid nanospheres which have limited electrochemical performances, hollow nanospheres have gained extensive interest. For instance, Gao et al. (2014) reported carbonaceous hollow spheres with a huge specific surface area (2,106.0 m^2/g) based on fermented rice. At a high current density of 15 A/g, this materials exhibited 219 F/g specific capacitance (Gao et al., 2014).

2.2 CNTs and other 1D carbonaceous materials

On account of their structural alignment, CNTs can be divided into two groups: single-walled (SWCNTs) and multi-walled carbon nanotubes (MWCNTs). A SWCNT is composed of a cylindrical-shaped graphene sheet with uniphase and outer phase bonds of sp2 carbon atoms (Haddon, 2002). The MWCNTs is generated by the co-dimensional arrangement of the SWCNTs (Baig et al., 2016). Kaskela et al. (2010) synthesized SWCNTs *via* thermal decomposition of ferrocene vapor in the atmosphere of carbon monoxide. Liu et al. (2011) pyrolyzed polypropylene into MWCNTs and hydrogen-rich gas. Balboni et al. (2022) prepared vertically aligned CNTs arrays (ACNTA) through photothermal chemical vapour deposition method, and ACNTA-polyaniline/polydimethylsiloxane electrodes were fabricated for a flexible supercapacitor which showed good cyclic stability and electrochemical properties. Its capacitance was able to maintain 80% at different bending angles (Balboni et al., 2022).

Carbon nanofibers (CNFs) are another kind of widely studied 1D carbonaceous materials. One of the most common procedure to synthesize CNFs is electrochemical spinning. Tao et al. (2019) synthesized electrospun CNFs based on waste walnut shells *via* the procedure of liquefying, electrospinning and carbonizing. Singh et al. (2021) obtained self-standing CNFs at different temperatures, they found the 800°C heat treated CNF exhibits largest specific capacitance (196.63 F/g at a current density of 1 A/g).

2.3 Graphenes and other 2D carbonaceous materials

Graphene is like a special kind of graphite, but only a single layer atomic thick. Pure graphene is a carbonaceous material with excellent electrical and thermal conductivity, strong physical strength and good chemical stability. Embed these properties into supercapacitors with good cyclic stability and high power density would be advantageous. Many nanocomposites with heteroatoms or functional groups can be formed based on graphene (Olabi et al., 2021). Wang et al. (2009) prepared graphene materials from graphene oxide sheets that synthesized through modified Hummers' method (Hummers and Offeman, 1958; Becerril et al., 2008) using hydrazine reduction to recover the conductive carbon network. They fabricated supercapacitors based on these graphene materials with maximum energy density of 28.5 Wh/kg, power density of 10 kW/kg and specific capacitance of 205 F/g at the current density of 100 mA/g (Wang et al., 2009). Long et al. (2015) presented the preparation of densely layer-stacking porous carbon nanosheets *via* hydrothermal treatment of the fungus and the later carbonization process. These 2D materials have 1,103 m^2/g surface area, and exhibit a large volumetric capacitance of 360 F/cm^3 at 200 mV/s scan rate (Long et al., 2015).

2.4 Carbon aerogels, CDCs, ACs and other 3D porous carbonaceous materials

Carbon aerogels usually can be obtained from organic aerogel through pyrolysis process. These highly porous carbonaceous materials exhibit good thermal, electrical and mechanical

properties, and has greater compressibility and adsorption. Structure and properties of carbon aerogels can be controlled by different preparation methods, which let carbon aerogels become one of potential candidate materials for supercapacitors (Pekala, 1989; Conway et al., 1997). Liang (2015) investigated the impact of heating rate and pyrolysis temperature on carbon aerogels' morphologies and electrochemical properties. It was found that carbon aerogels prepared by carbonization in nitrogen at 850°C have large specific capacitance, and the cyclic voltammetry curves are close to an ideal rectangle, with good capacitance characteristics and reversibility (Liang, 2015).

CDCs are a late-model type of porous carbonaceous type of carbon materials obtained through taking carbide as a precursor and trying to remove metal atoms or non-metal atoms in the lattice layer by layer, leaving a skeleton carbonaceous structure (Meng et al., 2004). By adjusting the temperature, atmosphere composition, reaction time and other parameters during the reaction process, nano-scale regulation can be achieved to obtain CDCs coatings with different structures (Chmiola et al., 2010). CDCs have become a kind of potential electrode materials for supercapacitor (Chmiola et al., 2010; Christians et al., 2021). Recently, Pang et al. (2021) obtained CDC@NC using polydopamine as the nitrogen and carbon source. The specific surface area of CDC@NC was up to 1,191 m^2/g. As a electrode material, the specific capacitance of CDC@NC reached 255 F/g at 0.5 A/g, and 193 F/g at 20 A/g (Pang et al., 2021).

ACs are prepared from carbon-containing raw materials through pyrolysis and activation processing. These carbonaceous materials usually have developed pore structures and high surface area (Nizam et al., 2021). Many supercapacitor electrodes based on ACs exhibit excellent performances not only owing to their highly porous structure but also due to their redox behavior (Boulanger et al., 2021; Rashidi et al., 2022). Recently, AC materials have been prepared from different biomass materials (e.g., coconut shell, cotton stalk, algae, saussurea involucrata stalk) (Sun et al., 2013; Gao et al., 2014; Tian et al., 2017; Wang et al., 2017; Wang et al., 2019). Elanthamilan et al. (2022) reported a mesoporous ACs made through chemical activation of dried fruit biomass at various temperatures. Their further fabricated symmetric supercapcitor exhibited an remarkable 11.11 Wh/kg energy density with 333.3 W/kg power density (Elanthamilan et al., 2022).

Carbon foams, which could possess good thermal stability, high mechanical strength, open cell structure, adjustable electrical and thermal conductivities, are mainly prepared *via* three routes: Compaction and exfoliation of graphite, template carbonization of carbon precursor as well as blowing of carbon precursors such as pitches with subsequent carbonization (Li et al., 2011). Hierarchical carbon foams with suitable surface functional groups and considerable surface area are good candidates for advanced supercapacitor electrode materials (Lv et al., 2012; Yang et al., 2022b). Xiao et al. (2016) reported a highly compressible, hydrophilic and free-standing N-doped carbon foam which could not only endure a compressive strain of 80% but also achieve 343 mF/cm^2 and 52 F/g specific capacitance at 1 mA/cm^2 current density.

3 Comparisons of different carbonaceous electrode materials

Comparisons of different carbonaceous materials for supercapacitor are listed in Table 1. From the view of industrial production, although aerogel, CDCs, CQDs, CNTs, and graphene materials exihited controllable pore structures, large surface area and high chemical stability, all these materials, which may be good candidates for preparing on-chip microsupercapacitors, require time-consuming, complicated and expensive producing process, which probably hinder their large-scale application. On the other hand, ACs have been reported as the most preferable carbonaceous electrode materials (Wang et al., 2015). To further reduce the cost of industrial production of carbonaceous electrode materials, low cost precursors should be preferred. Carbonaceous wastes such as plastic wastes, agricultural biomass wastes, industrial wastes, medical wastes and municipal wastes have great recycling value. Rational design of chemical processes for converting carbonaceous waste into carbonaceous electrode materials can also reduce potential carbon dioxide emissions (Tatrari et al., 2021). On the other hand, it is essential to probe new methods to further enhance electrochemical performances of carbonaceous electrode materials.

4 Functionalization of carbonaceous electrode materials

The nature of carbonaceous electrode materials can be monitored and adjusted through heteroatom doping and surface functionalization with different methodologies. For instance, the position and performances of surface functional groups can be adjust *via* thermal heating or chemical treatments. On the other hand, doping of heteroatoms (e.g., O, N, F, S, B, P and metallic atoms into carbon skeleton will often adjust carbonaceous materials' wettability, electroconductibility and capacitance. As shown in Supplementary Figures S7, S8, those surface functionalization can be either derived from heteroatoms containing precursors or achieved through post-treatment/activation process (Teimuri-Mofrad et al., 2022; Lachawiec et al., 2005; Maldonado-Hódar et al., 2000; Ji et al., 2009; Ramirez et al., 2021; Liu et al., 2017; Qin et al., 2009; Mei, 2022; Ping et al., 2016; Zhai et al., 2021; Hoa et al., 2021; Chen et al., 2021; Cao et al., 2021; Zhou et al., 2021; Wang and Pumera,

TABLE 1 Comparisons of different carbonaceous electrode materials for supercapacitors.

Carbonaceous materials	Carbon aerogels	CDCs	Carbon foams and templated carbon	Graphene and graphene derived materials	CQDs, CNTs and their derived materials	ACs
Preparation cost	Medium (M)	M	M	M	High (H)	Low (L)
Scalability	M	M	M	M	L	H
Conductivity	L	M	M	H	H	L
Specific surface area (m^2/g)	≤1,000	≤3,500	≤4,500	2,630 (Theoretical values for graphene) Rashidi and Yusup (2020)	1,315 (Theoretical values for CNTs) Rashidi and Yusup (2020)	≤3,000

2016; Mousavi-Khoshdel et al., 2016; Mousavi-Khoshdel and Targholi, 2015; Silva et al., 2020; Song et al., 2018; Morteza et al., 2015; Kolosov and Glukhova, 2021a; Kolosov and Glukhova, 2021b; Song et al., 2021c; Vermisoglou et al., 2021; Tong et al., 2021; Chakrabarty et al., 2021; Liu et al., 2022; Paul1 and Roy, 2021; Basivi et al., 2021; Zhang et al., 2019; Zhai et al., 2011; Rashidi and Yusup, 2020; Yang and Zhou, 2017; Arango et al., 2018; Umezawa et al., 2021; Mandal et al., 2021; Wulandari et al., 2021; Wu et al., 2021; Taer et al., 2021; Kalu-Uka et al., 2022; Yuan et al., 2021; Gopalakrishnan and Badhulika, 2020; Wang et al., 2015; Du et al., 2021; Jin et al., 2019).

4.1 Functionalization of carbon aerogels

Modification of aerogels is a major trend for carbon aerogels. At present, the modification methods of aerogels mainly include surface modification, doping and composite (Lachawiec et al., 2005; Teimuri-Mofrad et al., 2022). Maldonado-Hódar et al. (2000) discovered that the Co, Ni, doped carbon aerogel system undergoes homomorphic structural reorganization, from the original nano-continuous network structure to the nano-graphite structure. Ji et al. (2009) reported a Ni^{2+} doped carbon aerogel, which has specific surface area of 441.8 m^2/g. At 50 mA/g current density, this material achieve 227.3 F/g specific capacitance, which is higher than that of pure carbon aerogels (93 F/g) and Fe^{3+} doped carbon aerogel (180.5 F/g) (Ji et al., 2009). Ramirez et al. (2021) fabricated carbon aerogels with various Pd contents. It was found that Pd doping resulted in significantly enhanced power and energy density compared to undoped carbon aerogels. Liu et al. (2017) incorporated the surface of carbon aerogel with size-selected and dispersed WO_3 nanoparticles using solvent-immersion process and calcination treatment. This modified carbon aerogels show 1055 F/g specific capacitance at 5 mV/s scanning rate (Liu et al., 2017). Qin et al. (2009) prepared a nanocomposite named ANC/ACA which is composed of carbon aerogel (ACA) and activated nitrogen-enriched carbon (ANC). At the scanning rate of 1 mV/s, ANC/ACA with composite ratio of 12:1 exhibits 312.8 F/g specific capacitance, higher than that of ACA (103.4 F/g) and ANC (230.1 F/g) (Qin et al., 2009).

Conventional carbon aerogels based on phenolic resins are expensive and generate toxic pollutants during pyrolysis. Recently, several biomass carbon aerogels have been developed. Biomass resources are usually rich in oxygen and nitrogen elements, these characteristics could be used for self-activation to achieve the effect of self-doping (Mei, 2022). Ping et al. (2016) reported a carbon fiber aerogel based on natural cotton. The specific surface area of these carbon aerogels can be adjusted from 1,536 to 2,436 m^2/g *via* controlling the KOH amount in activation process (Ping et al., 2016). Zhai et al. (2021) reported a series of N self-doped carbon aerogel (NSCA) using chitin as carbon and nitrogen sources through microwave hydrothermal reaction and carbonization with Zinc chloride as the activating and dehydrating agent. NSCA-1000 exhibited 249.4 and 164.9 F/g specific capacitances at the current densities of 1.0 and 10 A/g (Zhai et al., 2021). Hoa et al. (2021) combined reduced graphene oxide (RGO) with $NiCo_2S_4$ to prepare RGO/$NiCo_2S_4$ aerogel, which have good electrical conductivity and high porosity. RGO/$NiCo_2S_4$ aerogel exhibit a capacitance of 813 F/g at 1.5 A/g current density. The asymmetric supercapacitor based on RGO/$NiCo_2S_4$ aerogel reaches 40.3 Wh/kg energy density with 375.0 W/kg power density (Hoa et al., 2021). Chen et al. (2021) synthesized S- and N-doped 3D porous graphene aerogels (NS-3DPGHs) with thiourea as a reducing agent and additive. The NS-3DPGH-150 sample exhibits 412.9 F/g specific capacitance with remarkable cycling stability at 0.5 A/g current density (Chen et al., 2021). Cao et al. (2021) Reported N, Fe co-doped (FeNC) aerogel, which exhibit up to 3,103 m^2/g specific surface area and 170–264 F/g specific capacitances at the current density of 0.5 A/g. With the increase of iron content, the cycling stability and capacitance performance of FeNC aerogel was decreased (Cao et al., 2021). Zhou et al. (2021) developed a facile way to combine carbon aerogels with MnO_x nanoparticles and prepare carbon aerogel/MnO_x composite. As shown in Supplementary Figure S3, MnO_x nanoparticles grow consistently on carbon aerogel surface. The uniform growth of MnO_x nanoparticles and cross-linked 3D

structure of carbon aerogel are advantageously to the composite's electrochemical performances. Specific capacitance of carbon aerogel/MnO_x reached 557 F/g at 1 A/g current density (Zhou et al., 2021).

4.2 Functionalization of low-dimensional carbonaceous electrode materials

Both CNTs and Graphene are low-dimensional electrode material for supercapacitor. Theoretically, C_Q of CNTs graphene maybe improved by suitable functionalization (Mousavi-Khoshdel and Targholi, 2015; Mousavi-Khoshdel et al., 2016; Wang and Pumera, 2016). Silva et al. (2020) employed density functional theory (DFT) calculations and molecular dynamics atomistic simulations to determine C_T. Their results exhibited that doped electrodes present variations in C_Q which is in the range of 0–200 μF/cm^2, because of the changes in the pristine graphene's electronic structure. However, neither vacancies nor doping atoms has appreciable influence on C_D (Silva et al., 2020). Song et al. (2018) explored the C_Q of graphene oxide (GO) through DFT calculations. As shown in Supplementary Figure S4, it was found that intoducing oxygen-containing groups can enhance the total density of states near the Fermi level. Moreover, increasing the concentrations of hydroxy and epoxy groups will improve C_Q of GO (Song et al., 2018).

Mousavi-Khoshdel M. investigated the impact of doping (P, S, Si) and co-doping (P N, S N, Si N) of graphene on structural parameters, electronic properties as well as C_Q *via* DFT and generalized-gradient approximation (GGA). The obtained results indicated that in comparison with pristine graphene sheets, codoping of graphene generates new electronic states and accumulates larger quantity of electrical charge on co-doped graphene sheets, which will enhance their quantum capacitance consequently (Morteza et al., 2015). As shown in the Supplementary Figure S5, Kolosov and Glukhova (2021a) predicted ways to improve the performances of the electrode material on the base of pillared graphene (PGR) and boron clusters B_{12} through first-principle DFT method. Specific charge density calculation proved the symmetry of positive and negative bias at 5.13 wt. % (two B_{12} clusters). Moreover, branch asymmetry was observed when the increase in boron concentration, therefore during the addition of B_{12} clusters, the electronic states density is increased, the electrode conductivity will probably be increased. The electronic properties and C_Q of SWCNTs decorated with B_{12} clusters were also explored. As shown in Supplementary Figure S6, it was found that the addition of B_{12} clusters can improved the C_Q of SWCNTs (Kolosov and Glukhova, 2021b).

Numerous reported experimental studies also demonstrated that functionalizing graphene and carbon nanotubes can effectively improve their electrochemical performances. Song et al. (2021c) synthesized N-doped graphene sheets (NGS) through a solid-state microwave-mediated process. Subsequently, experimental electrodes employing NGS were fabricated. At 0.5 A/g current density, the electrode's specific capacitance reached 208.17 F/g (Song et al., 2021c). Vermisoglou et al. (2021) utilized fluorographene, which was uniformly linked with amino acid moieties on graphene's both sides. By adding non-toxic pore formers and adjusting conditions of reactions, the hierarchical porosity and optimal degree of functionalization were achieved. At 0.25 A/g current density, this material exhibited high capacitance of about 390 F/g. The assembled supercapacitor device was tested for 30,000 cycles of charge-discharge at a current density of 2 A/g, showing 82.3% capacitance retention (Vermisoglou et al., 2021). Tong et al. (2021) synthesized poly-3,4-ethylenedioxythiophene (PEDOT) on graphene nanosheet-deposited open-pored polyurethane (PU) sponge by vapor-phase polymerization (VPP) way to prepare porous electrodes. Under 0.1 mA/cm^2 current density, the obtained electrodes showed high areal specific capacitance of 798.2 mF/cm^2 (Tong et al., 2021). Chakrabarty et al. (2021) prepared RGO sheets of different weights decorated with CeO_2/Ce_2O_3 quantum dots ($CRGO_3$). In tests, $CRGO_3$ showed a capacitance of 1027 F/g at 1 A/g (Chakrabarty et al., 2021). Liu et al. (2022) prepared graphene hydrogel (GH) nanocomposites with graphitic carbon nitride (g-C_3N_4) quantum dots (CNQDs), in which GH are decorated with CNQDs through a 3D layered interconnected configuration. The obtained CNQ/GH nanocomposites exhibited 243.2 Fg^{-1} specific capacitance at 0.2 Ag^{-1} current density (Liu et al., 2022). Paul and Roy (2021) developed a spray pyrolysis process for the synthesis of radially grown nanoporous brush-like CNTs on carbon cloth (CC) substrates with boron (B) and nitrogen (N) simultaneously co-doped and a composite BNCNT-CC was obtained. Based on BNCNT-CC, highly flexible all-solid-state symmetric supercapacitors was further integrated, which have high capacitance retention (86.4%) after 5,000 charge-discharge cycles at 5 mA/cm^2 (Paul1 and Roy, 2021). Basivi et al. (2021) synthesized a novel composite, N-doped MWCNT/carboxymethyl cellulose (N-MWCNT/CMC) for electrodes of supercapacitors. The synthesized composite exhibited 274 F/g specific capacitance at 2 A/g. After 4,000 cycles of the composite electrode, the cycling stability is as high as 96%, which is beneficial for supercapacitor applications (Basivi et al., 2021).

4.3 Functionalization of ACs

Hydrophobic nature of unmodified ACs coursed weak infiltration of electrolyte to electrode, and ion-accessible surface limitation (Zhang et al., 2019). Zhai et al. (2011) concluded that porous structure, conductivity and surface

properties optimization are required to ensure maximum capacitance of ACs. Functionalization of ACs as supercapacitor electrode materials has attracted more and more attentions nowadays (Rashidi and Yusup, 2020) Yang and Zhou (2017) reported N atoms inclusion into the structure of ACs to provide an extra redox reaction for pseudocapacitor. Arango et al. (2018) reported a HNO_3 modified ACs with 1.8 wt. % N content, compared with only 0.4 wt. % of the unmodified AC. After 1,000 operating cycles at 0.27 A/g, this modified AC maintains 75.3% capacitance retention (Arango et al., 2018). Rashidi and Yusup (2020) sorted out recent researches about N modification on different ACs. From 2015 to 2020, nitric acid, ammonia and poly (ethylene imine) were reported as post-modification chemical agents to functionalize the surface of ACs. After functionalization, the capitance value of obtain nitrogen-functionalized ACs are at least doubled compare with unmodified AC. Melamine and urea are often used as *in situ* nitrogen doptant, and biomass raw materials (e.g., swim bladder, corncob, sakura et al.) can be used to prepare nitrogen-self doped ACs for supercapacitors (Rashidi and Yusup, 2020). Umezawa et al. (2021) prepared B-doped ACs based on B-based covalent organic frameworks (COF-5). The supercapacitor electrodes made from it show 15.3 μF/cm^2 specific capacitance at 40 mA/g, which is much larger than that of pristine ACs (Umezawa et al., 2021). Mandal et al. (2021) prepared a novel composite of AC/MWCNTs-$ZnFe_2O_4$, which exhibit 613 F/g specific capacitance at 5 mV/s scan rate, and after 10,000 cycles, 91% of the capacitance can be retained. Wulandari et al. (2021) used $CaCl_2$ as an active agent to prepare AC from fruit bunches, after carbonization and activation processes, the product was further modified with 1 mol/L nitric acid (HNO_3). Wu et al. (2021) developed N, O, S co-doped hierarchical AC, which has 2,864 m^2/g specific surface area with 1.461 m^3/g total pore volume. This material exhibits 270 F/g specific capacitance at 1 A/g with remarkable long-term cycling stability and 98% capacity retention after 10,000 cycles (Wu et al., 2021). Taer et al. (2021) fabricated a biochar AC using reproducible and environmentally friendly branch and leaf (Syzygiumoleana). The feedstock is converted into biochar AC using $ZnCl_2$ impregnation and one-stage integrated pyrolysis. This biochar AC provides 138.5 F/g specific capacitance at a low scan rate of 1 mV/s in 1 mol/L H_2SO_4 aqueous electrolyte (Taer et al., 2021). Kalu-Uka et al. (2022) convert termite biomass into AC using KOH as the activator through one-process carbonization activation procedure. The specific capacitance of the best AC is 91.76 F/g at 0.5 A/g in 1 mol/L H_2SO_4, while its specific capacitance achieved 62.35 F/g at 0.5 A/g in 1 mol/L $EMImBF_4$ (Kalu-Uka et al., 2022). Yuan et al. (2021) reported N-doped AC based on rice paddies using atmospheric pressure blasting, activation and N doping process. The prepared N-doped AC has maximum 417 F/g specific capacitance at 1 A/g (Yuan et al., 2021). Gopalakrishnan and Badhulika (2020) reviewed a self-doped heteroatom ACs for supercapacitors. The impacts of doping (N, S, P) on electrochemical performance were highlighted in their review. They concluded that N doped ACs are beneficial in improving conductivity and hydrophilicity. S doped ACs result in electronic reactivity enhancement of carbon and increase pseudocapacitive contribution. P doped ACs enlarge operating potential of cells thus increase energy density. Dual (N, S or N, P) or multi-heteroatom (N, S, P) doped ACs have synergistic effects of dopants which may yield superior performance (Gopalakrishnan and Badhulika, 2020).

5 Summary and future perspectives

This review proposes a brief description about the operating principle and evaluation parameters for supercapacitor. Since both EDLCs and pseudocapacitors occur on surface electrodes, carbonaceous electrode materials' surface strongly affect the electrochemical performances of supercapacitor, which could be further improved through heteroatom doping and surface functionalization. Theoretically, C_Q of CNTs and graphene could be improved *via* heteroatom doping, which were demonstrated by experimental studies. As a major trend for carbon aerogels and ACs, surface modification, doping and composite can not only improve their optimization of pore size and structure, conductivity and surface properties, but also introduce extra pseudocapacitance to these materials.

Different carbonaceous materials were roughly classified and their preparation process and properties was discussed briefly. Compared with other carbonaceous materials, ACs and aerogels from biomass have drawn more and more attention nowadays owing to its inexpensive cost and renewablity. Especially, the production and utilization of domestic, medical and industrial wastes residues derived carbonaceous materials should be of great significance for controlling environmental pollution and reducing CO_2 emissions, which we think will be worth to further studies.

Articles on the manufacturing technology and equipments for carbonaceous materials are relatively rare. Moreover, articles discussed waste gas composition and disposal methods in different carbon material industrialization processes (carbonization, activation) are very few. Which we think should be further study in future.

Author contributions

Conceptualization, YL and JW; methodology, XH and XS; investigation, DJ and SZ; writing-original draft preparation, YL,

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Conflict of interest

JL was employed by Hangzhou Plastics Industry Co., LTD.

The remaining authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

Publisher's note

Supplementary material

The Supplementary Material for this article can be found online at: https://www.frontiersin.org/articles/10.3389/fenrg.2022.957032/full#supplementary-material

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