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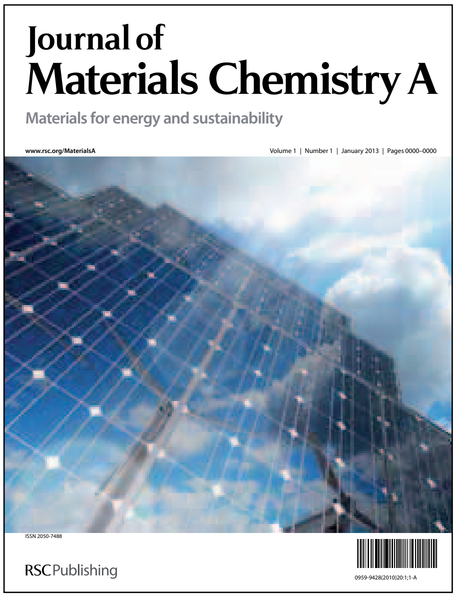

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FEATURE ARTICLE

Carbonaceous hydrogels and aerogels for supercapacitors

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Carbonaceous hydrogels and aerogels are a large class of gels which have received much attention due to their multifunctional properties. The three-dimensional networks and porous structure of the carbonaceous gels can provide efficient diffusion of electrolyte ions and electrons, leading to promising applications in supercapacitors. This feature article gives an overview of the recent advances of using novel carbonaceous gels for supercapacitors. In particular, the synthetic methods of polymer derived carbonaceous gels, carbon nanotubes based carbonaceous gels, graphene based carbonaceous gels and biomass derived carbonaceous gels are introduced, and their applications for supercapacitors are systematically discussed. Perspectives of the development of neotype carbonaceous gels based electrode materials for supercapacitors are given.

1 Introduction

There has been an increasing worldwide demand for the development of alternative energy techniques with more efficiency and sustainability due to the fast consumption of fossil fuels and related enviromental issues of climate change and global warming. Over the past decades, many efforts have been made in developing new techniques toward clean energy including biofuels, wind power and solar energy, as well as efficient energy storage such as lithium-ion batteries and supercapacitors.[1] As more and more renewable energy resources used, the development of high-performance, low-cost and environmental-friendly energy storage devices is crucial to pratical applications (such as consumer electronics, electric and hybrid vehicles) and the future sustainable economy. Among the various energy storage systems, supercapacitors, or electrochemical capacitors are considered as promissing alternative power source for rapid storage and release of electric energy as a result of their high energy density, simple principles, long cycle life, long-term operation stability, low level of heating and high rate capability.[2,3]

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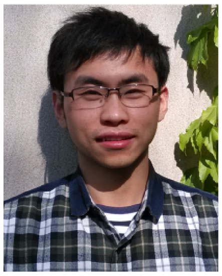

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A supercapacitor consists of two nonreactive porous plates, or electrodes immersed in an electrolyte and seperated by a porous membrane that allows migration of electolyte ions and electrically isolates the two electrodes. According to the charge storage mechanisms, supercapacitors can be classified into two types: pseudocapacitor and electric-double-layer capacitor (EDLC). The pseudocapacitors store energy via the fast *Faradaic* redox reactions of the electrode materials, while the electric-double-layer capacitors store energy via the adsorption of electrolyte ions on the surface of electrode materials.[1,3] Therefore, metal oxides and conducting polymers that can undergo reversible redox reactions are usually used as electrode materials for the pseudocapacitors. While materials with high specific surface area are beneficial for the adsorption of electrolyte ions, which are suitable for EDLC.

Carbonaceous materials with high surface area and porous structure have been widely investigated as electrode materials for supercapacitors (for example: activated carbons,[4] templated carbons[5] and carbon aerogels[6]). Carbonaceous hydrogels and aerogels are a large class of porous carbonaceous materials. Carbonaceous hydrogels possesss the characteristics of high water absorptivity, good elasticity and good osmotic property, which have been investigated for the applications as sensors,[7] adsorbents,[8] electrode materials for batteries and supercapacitors.[6,9] Carbonaceous aerogels exhibit the characteristics of high porosity, high surface area and low density, leading to promissing applications in the field of adsorbents,[10] electrodes[11] and catalyst support.[12] Carbonaceous gels manifest an open three-dimensional (3D) networks and porous structure, which can provide efficient diffusion/mass transfer of liquid/gas phase and large amount of active sites for the adsorption of ions/molecules.[6] The unique structure of the carbonaceous gels could allow stable and sustained transport of both electrolyte ions and electrons to the surface, leading to promissing applications for supercapacitors. On the other hand, the carbonaceous gels can serve as robust substrates, decorating metal oxides or conducting polymers, which can enhance the capacitance.

In this feature article, basic knowledge of the thoery for supercapacitors and backgrounds for the carbonaceous gels are introduced. Recent advances in carbonaceous gels are summarized, particularly in the related synthetic methods and their aplications for supercapacitors. In detail, the fabrication of polymer derived carbonaceous gels, carbon nanotubes based carbonaceous gels, graphene based carbonaceous gels and biomass derived carbonaceous gels are discussed. Various synthetic methods for the synthesis of carbonaceous gels including freeze-drying, hard template, soft template and hydrothermal treatment are discussed. Applications and perspectives of the above carbonaceous gels as electrolyte or electrode materials are discussed in detail.

2 Backgrounds for supercapacitors, hydrogels and aerogels

2.1 Basic knowledge for supercapacitors

Depending on their structure and properties, carbonaceous gels work mostly as electric-double-layer capacitor (EDLC) for supercapacitors. Fig. 1 shows the structure of EDLC containing two electrodes inmmersed in an electrolyte and a separator located between the two electrodes. The main mechanisms for the EDLC arise from the reversible adsorption of electrolyte ions on the surface of the electrode materials. During the charge process, anions transport to surface of positive electrode and cations transport to negative electrode, forming electrical double layers at the interface. While the stored energy released, the adsorbed electrolyte ions will release to the solutions. The accumulated ions on the electrodes result in a capacitance C as described by Helmholtz[13] by the following formula:

$$C = \frac{\varepsilon_r \varepsilon_0 A}{d} \qquad (1)$$

where ε_r and ε_0 are the electrolyte and vacuum dielectric constants, respectively, d is the effective thickness of the electrical double layer, and A is the surface area of the interface. Each electrode (C_1 or C_2) can be considered as a capacitor. The total capacitance of a supercapacitors cell (C_{cell}) can be calculated by using Equation 2:

$$\frac{1}{C_{cell}} = \frac{1}{C_1} + \frac{1}{C_2} \qquad (2)$$

where C_1 and C_2 are the capacitance of the positive and negative electrodes, respectively. The electric energy (E) of the

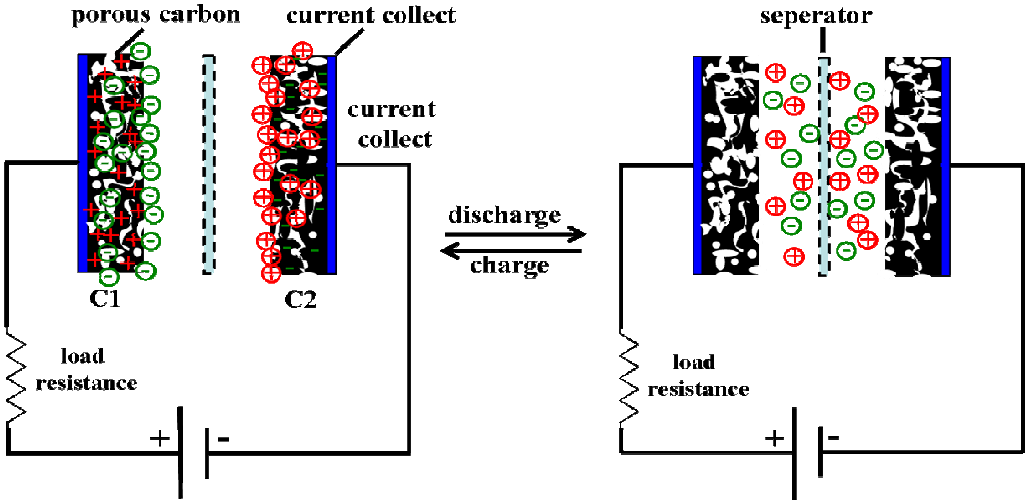

Fig. 1 Schematic diagram of the charged and discharged electric double layer capacitor.

supercapacitor is related to the capacitance (C) and the applied voltage (V):

$$E = \frac{1}{2}CV^2 \qquad (3)$$

The capacitance is essentially depended on the electrode materials used. The use of high-capacitance materials is crucial to the obtaining of high electric energy and power density. The power (P) of a supercapacitor can be calculated by following equation:

$$P = \frac{V^2}{4R_s} \qquad (4)$$

where V is the maximun cell voltage and R_s is the the equivalent series resistance (ESR) in ohms. The capacitance of EDLC is mainly dependent on the characteristic of the electrode materials such as conductivity, surface area and pore size distribution. For further improve the capacitance, other materials such as metal oxides or conducting polymers that exhibit pseudo-capactive behavior can be doped in the electrode materials.

2.2 Backgrounds for carbonaceous hydrogels and aerogels

Hydrogels are physically or chemically cross-linked 3D hydrophilic solid networks with adsorbed water in the networks. As one of the soft materials, hydrogels have been widely applied as artificial tissues,[14,15] drug carriers,[15] electrode materials,[6,16] actuators,[17] sensors,[18] and adsorbents.[19] The classification of the hydrogels are dependent on a variety of characteristics such as the source, the nature of cross-linking (covalent or non-covalent) and the structural features (affine or phantom).[20] Traditional hydrogels crosslinked by organic polymers are usually associated with problems of poor mechanical properties and stability which limits their applications. Novel hydrogels or hybrid hydrogels with hierarchical structures and multifunctional properties have been synthesized by using advanced materials (carbon nanotubes, graphene, biomass etc.) as constituent building blocks or incorporating of other functinal materials such as nanoparticles into the hydrogels. For example, multifunctional hydrogels or hybrid hydrogels including carbonaceous nanofiber hydrogels,[8] graphene–poly(N,N-dimethylacrylamide) composite hydrogels,[14] graphene hydrogels,[16,17] and polymer–clay nanosheet composite hydrogels[21] have been successfully fabricated recently. Novel hydrogels with inorganic or organic-inorganic hybrid building blocks have drawn many attentions due to their excellent mechanical properties. Our group[6] has fabricated carboncaeous hydrogels and aerogels by using crude biomass as the source and demonstrated their robust mechanical properties (**Fig. 2**). As can be seen in **Fig. 2a**, the carboncaeous hydrogels can be easily compressed in water and can recover its original scale. The compressive stress–strain curve of the hydrogels shows the maximal stress of 23.8 KPa at a set stain (ε) of 60% (**Fig. 2c**). The dynamic viscoelastic measurements (**Fig. 2b**) at angular frequencies of 1–100 rad·s^{-1} show storage modulus of about 948.5 KPa at 10 rad·s^{-1} and the storage modulus value is much higher than the loss modulus value over the entire angular frequencies, indicating an excellent elastic

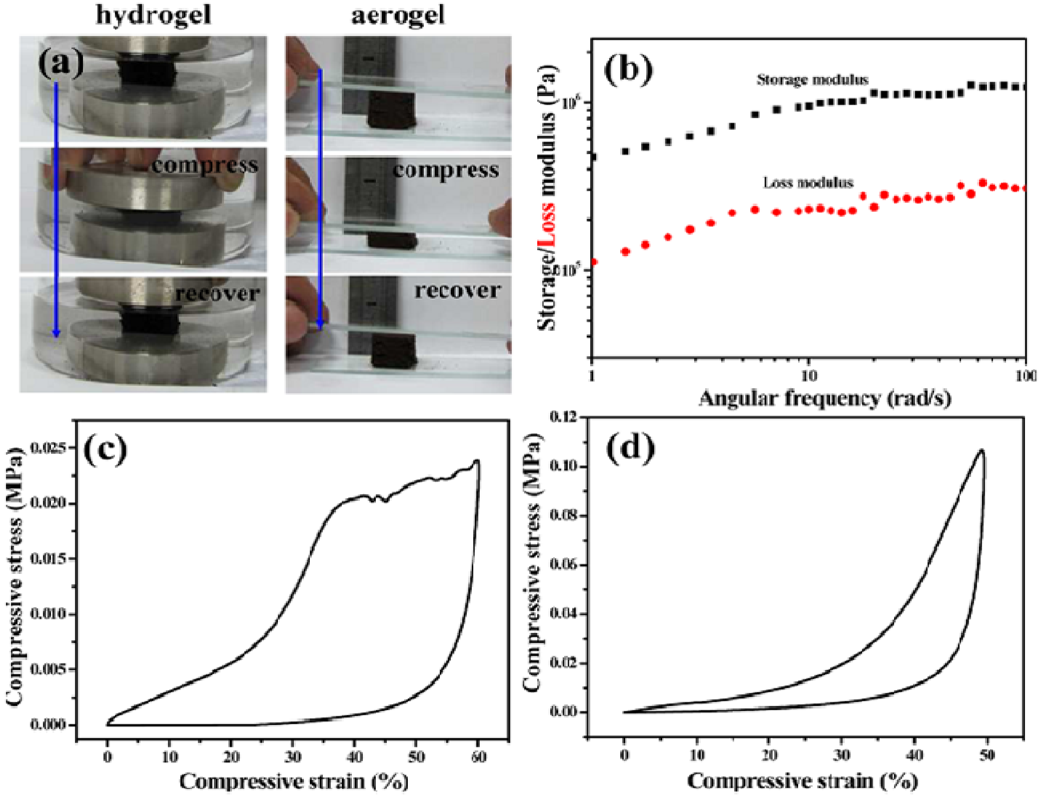

Fig. 2 (a) Digital pictures show the compressive properties of carbonaceous hydrogel and aerogel. (b) Dynamic rheological behavior of carbonaceous hydrogel. (c) Compressive stress–strain diagrams of the hydrogel at ε = 60% and (d) Compressive stress–strain diagrams of the aerogel at ε = 50%.

property of the carbonaceous hydrogels.

Aerogels have a similar structure to that of hydrogels. Aerogels can be synthesised by replacing the liquids in hydrogels or other wet gels by air without collapsing the interconnected microstructure. The properties of low density (0.004–0.500 g·cm^{-3}), high surface area and large open pores of the aerogels could lead to many advanced applications such as supercapacitors,[6,22] electrocatalysis,[23] adsorbents,[8,24] stretchable conductors,[25,26] lithium-ion batteries,[27] and artificial muscles.[28] The traditional aerogels usually associate with problems of poor mechanical strength and using supercritical drying which is less energy efficient. Thus, many efforts have been made aiming to synthesis functional aerogels with strong mechanical properties and, more importantly, to develop novel and cost-effective synthetic methods. For example, novel aerogels based on carbon nanofiber,[24] graphene,[26] nanocellulose,[29] and epoxy-clay[30] are fabricated by using freeze-drying methods. Wei *et al.*[31] prepared silica-polymer composite aerogels by a simple sol-gel process stepped by air-drying. The obtained composite aerogels showed properties of low high-temperature thermal conductivities and high mechanical strength. Novel carboncaeous aerogels fabricated by our group[6] by using a freeze-drying method exhibited an excellent compression properties as shown in **Fig. 2a** and **Fig. 2d**.

Carbonaceous materials based hydrogels and aerogels not only inherit the advantages of the gels but also exhibit some unique properties such as high electrical conductivity, high thermal conductivy and high biocompatibility. The high surface area and high electrical conductivity of the carbonaceous gels offer a great potential for the application in supercapacitors. More importantly, a growing demand for wearable and miniaturized electronic devices require the development of high-performance, lightweight and wearable energy convertion and storage systems. Thus, the carbonaceous gels based supercapacitors could be a suitbale choice for the power supply due to their excellent properties of high performance, lightweight, flexibility and stretchability.

3 Carbonaceous gels for supercapacitors

Over past several decades, many efforts have been made in synthesizing novel carbonaceous hydrogels and aerogels for supercapacitors. On the basis of the source, the carbonaceous gels can be classified into four groups: (1) polymer derived aerogels, (2) carbon nanotubes based aerogels, (3) graphene based hydrogels and aerogels, and (4) biomass-derived hydrogels and aerogels. Some recent advances focus on the development of carbonaceous hydrogels and aerogels based supercapacitors with characteristics of large capacitance, light-weight and flexibility. For further improving the capacitive properties of the carbonaceous gels, redox-reaction materials such as conducting polymers and metal oxides can be introduced into the networks of the gels to form a composite material.

3.1 Polymer derived aerogels for supercapacitors

Polymer derived carbon aerogels (CAs) are large class of carbonaceous aerogels. In 1989, Pekala and Kong first reported the synthesis of CAs by pyrolyzing resorcinol–formaldehyde (RF) organic aerogel in an inert atmosphere.[32,33] The RF organic aerogel are prepared by the sol–gel poly-condensation of resorcinol and formaldehyde monomers in water and subsequent by supercritical drying.[34] Other sol–gel derived polymers are also employed as precursors for preparing CAs, including phenol–melamine–formaldehyde gel,[35] poly-benzoxazine gel,[36] cresol–resorcinol–formaldehyde gel,[37] resorcinol–pyrocatechol gel,[38] cresol–formaldehyde gel,[39] resorcinol–methanal gel,[40] and poly(vinyl chloride) gel.[41] In addition to supercritical drying, microwave drying,[42] ambient drying (or air drying),[43,44] and freeze-drying[45] are also applied for the preparation of dried organic gels. The dried organic gel precursors can be transformed into CAs by pyrolysis. The structure of the CAs can be easily adjusted by controlling the composition of the precursors and the pyrolysis conditions (pyrolysis temperature and time).

Polymer derived CAs has been studied extensively as electrode materials for supercapacitors. Pekala first demonstrated that CAs have great potential for the application as electrodes in supercapacitors in 1994.[46] The 3D CAs are composed of interconnected nanoparticles (3 – 30 nm) with a large number of interstitial pores (< 50 nm) created by the interconnected solid networks. CAs hold the advantages of high porosity (80–98%), high electrical conductivity (25 – 100 S·cm^{-1}), high surface area (> 400 m^2·g^{-1}), controllable pore structure and low electrical resistivity.[46,47] Benefiting from their unique structure and excellent properties, CAs offer very attractive prospects for applications as electrodes in supercapacitors. Pekala *et al.* studied the synthesis, structure–property relationships of CAs for supercapacitors.[48] The effects of aerogel density, CO$_2$ activation, pyrolysis temperature and Ru-doping on capacitance values of the CAs were investigated. They demonstrated that the capacitance properties of the CAs are dependent on the structure and properties of CAs, which can be adjusted by controlling the parameters such as the ratio of the organic precursor, dopant compounds and pyrolysis temperature. Some strategies have been developed to enhance the performance of the CAs based supercapacitors, including activation, doping with metallic compounds, and combined with conducting polymers or other materials. Liu *et al.* prepared CO$_2$ and KOH activated CAs and investigated their performanc for supercapacitors.[49] The activated-CAs exhibit hierarchically porous structure with high specific surface area. There are three types of pores in the activated CAs: micropores (< 2 nm), small mesoporous (2 nm ~ 4 nm) and large pores (> 30 nm). The KOH activated CAs possesspore volume of 2.73 cm^3·g^{-1} and very large surface area of 2119 m^2·g^{-1}. Meanwhile, the KOH activated CAs exhibit a high capacitance of 250 F·g^{-1} and power density of 8.49 Wh·kg^{-1} at current density of 0.5 A·g^{-1} in 6 M KOH aqueous solution. The high capacitance of the activated CAs supercapacitors could be ascribed to the large surface area and the presence of a large number of micropores, which is beneficial to charge separation and transport.

Other porous carbons used in supercapacitors such as activated carbons and templated carbons are usually asociated with problems of low efficiency. Athough activated carbons have a high specific surface area of about 1000–2000 m^2·g^{-1}, there are not enough mesopores for the tranportation of

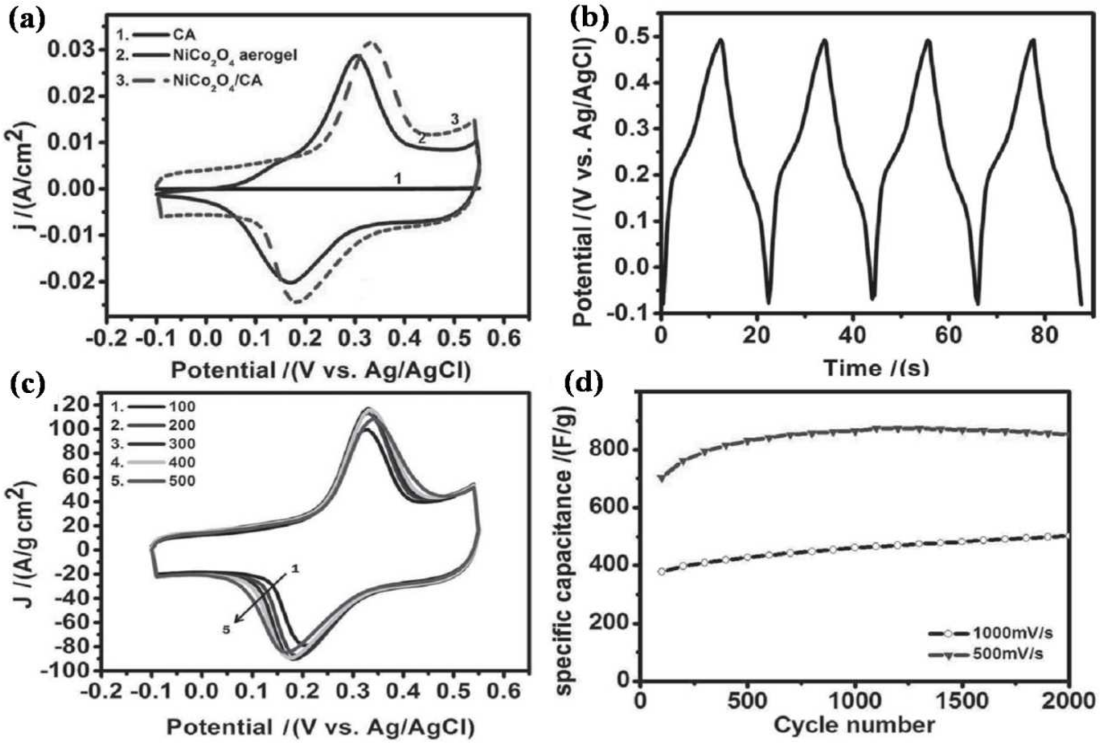

Fig. 3 (a) Representative cyclic voltammetry (CV) curves of the CA, $NiCo_2O_4$ aerogel and $NiCo_2O_4$-CA composite. (b) Charge/discharge curves of the $NiCo_2O_4$-CA composite. (c) CV curves of the $NiCo_2O_4$-CA composite based electrode at different scan rates. (d) Cycle lifes of the $NiCo_2O_4$-CA composite based electrode at a scan rate of 500 mV·S^{-1} and 1000 mV·S^{-1}, respectively.

electrolyte ions, which results in a low electrolyte accessibility. The mismatch of the ion size of the electrolyte and pore size of the electrode material is responsible for the low efficiency of activated carbons for supercapacitors. Similarly, templated carbons have highly ordered pores with consistent orientations and narrow pore size distribution. However, the single type of pores in the templated carbons are not good enough for the transportation of electrolyte ions. Moreover, the pores in templated carbons are usually accessible by a single channel, while the pores in CAs are all round and interconnected. The interconnected pores in CAs are much easier for the the tranportation of electrolyte ions. Some researchers suggested that an appropriate pore size is more important for achieving high specific capacitance than high surface area.[49] In addition, large mesopores and macropores are crucial for EDLCs at high charge/discharge current rate.[49] Thus, the high capacitive performance of CAs electrodes could be due to the presence of all the micropores, mesoporous and macropores. However, traditional CAs are usually associated with problems of poor mechanical properties which hinder them for the applications in flexible supercapacitors. There are also some other disadvantages of using CAs as electrode in supercapacitos such as difficulty for preparation and use of expensive raw materials.

Doping CAs with metallic compounds is demonstrated to be an efficient approach for enhancing the capacitance of carbon aerogels. Lee *et al.* fabricated a series of metal doped CAs and applied as electrode materials for supercapacitors.[50] They found that the metal doped CAs show higher capacitance than that of the pure CAs. The Mn-doped CAs exhibited the highest capacitance of 107 F·g^{-1}, while Cu- and Fe-doped CAs showed the most stable cyclability. Other metallic compounds doped CAs are also studied as elecreode materials for supercapacitors, inculuding Mn_3O_4-CAs,[40] $NiCo_2O_4$-CAs,[51] ZnO-CAs,[52] FeO_x-CAs,[53] MnO_2-CAs,[54] SnO_2-CAs,[55] NiO-CAs,[56] and RuO_2-CAs.[57] Chien *et al.* reported $NiCo_2O_4$ doping can significant enhance the capacitance of CAs.[51] **Fig. 3a** shows the representative cyclic voltammetry (CV) curves of the CA, $NiCo_2O_4$ aerogel and $NiCo_2O_4$-CA composite. The area of CV curve of $NiCo_2O_4$-CA composite is larger both than that of CA and $NiCo_2O_4$ aerogel, indicating an enhanced capacitance of the compoistes. The $NiCo_2O_4$-CAs based supercapacitor show ultrahigh specific capacitance of ~1700 F·g^{-1} at a scan rate of 25 mV·s^{-1} at a potential window of − 0.05 to 0.5 V in 1 M NaOH aqueous solutions. The charge/discharge curves of the $NiCo_2O_4$-CA composite show a symmetric shape with no obvious internal electric resistances (IR drops) (**Fig. 3b**). The capacitance obtained from the discharge curves reach a maximum of 1455 F·g^{-1} after 600 cycles. CV curves of the $NiCo_2O_4$-CA composite at scan rates of 100, 200, 300, 400 and 500 mV·S^{-1} display similar shapes, demonstrating an excellent cycling stability (**Fig. 3c**). Cycle life test also demonstrate the $NiCo_2O_4$-CAs supercapacitors have an excellent high rate capability by achieving a high specific capacitances above 800 F·g^{-1} at a high scan rate of 500 mV·s^{-1} and an outstanding cycling stability with retention about 97.6 % of the original capacitance after 2000 cycles charge/discharge test (**Fig. 3d**). The high performance of the composite materials could be due to the fuller utilization of nickel cobaltite for pseudocapacitance generation.[51] The composite structure could enable well exposure of nickel cobaltite to the electrolyte and ease for the

Table 1 Characteristics of various carbon aerogels based supercapacitors.

precursor	modification	surface area and pore size	capacitance	testing conditions	electrolyte	Ref.
polybenzoxazine	-	368 m2·g^{-1} 2-5 nm	55.78 F·g^{-1}	5 mA·cm^{-2} two electrode	3 M H_2SO_4	36
resorcinol–pyrocatechol	-	519 m^2·g^{-1} -	175 F·g^{-1}	0.1 A·g^{-1} three-electrode	1 M H_2SO_4	38
cresol–formaldehyde	CO_2 activation	1418 m^2·g^{-1} 11.9 nm	146 F·g^{-1}	1 mA·cm^{-2} three-electrode	30% KOH	39
resorcinol–methanal	Mn_3O_4 doping	577 m^2·g^{-1} 18 nm	503 F·g^{-1}	25 mV·s^{-1} three-electrode	0.5 M Na_2SO_4	40
resorcinol–formaldehyde	-	706 m^2·g^{-1} 10.9 nm	81 F·g^{-1}	10 mV·s^{-1} three-electrode	1 M H_2SO_4	44
resorcinol–formaldehyde	KOH activation	2119 m^2·g^{-1} 4.8 nm	250 F·g^{-1}	0.5 A·g^{-1} three-electrode	6 M KOH	49
resorcinol–formaldehyde	Mn doping	681 m^2·g^{-1} 8.7 nm	107 F·g^{-1}	10 mV·s^{-1} three-electrode	1 M H_2SO_4	50
resorcinol–formaldehyde	$NiCo_2O_4$ doping	206 m^2·g^{-1} 16.7 nm	1700 F·g^{-1}	25 mV·s^{-1} three-electrode	1 M KOH	51
resorcinol–formaldehyde	MnO_2 doping	120 m^2·g^{-1} 5 nm	515.5 F·g^{-1}	20 mV·s^{-1} three-electrode	1 M Na_2SO_4	54
resorcinol–formaldehyde	NiO doping	- -	356.2 F·g^{-1}	1 A·g^{-1} three-electrode	6 M KOH	56
resorcinol– formaldehyde	PANI modifying	13.4 m^2·g^{-1} 1.4 nm	710.7 F·g^{-1}	1 mV·s^{-1} three-electrode	1 M H_2SO_4	58
resorcinol– formaldehyde	CNTs modifying	- -	141.3 F·g^{-1}	5 mV·s^{-1} three-electrode	30% KOH	59

transportation of charge carriers, ions, and electrons. Another method available to enhance the performance of CAs based supercapacitors is to modify the CAs by conducting polymers. An *et al.* fabricated polyaniline/CAs composite materials by deposition of polyaniline polymers on the surface of CAs.[58] The obtained polyaniline/CAs composite was applied as electrode materials for supercapacitors, achieving a high specific capacitance of 710.7 F·g^{-1}, which is much higher than that of pure CAs (143.8 F·g^{-1}). Lv *et al.* fabricated CNTs modified CAs for supercapacitors and obtain a specific capacitance of 141.3 F·g^{-1} at 5 mV·s^{-1}, which is 1.4 times higher than that of the original CAs.[59] The studies on CAs based supercapacitors are summarized in **Table 1**, where the synthetic methods and the capacitance are compared.

3.2 Carbon nanotubes based aerogels for supercapacitors

CNTs aerogels are 3D porous networks that build up by using CNTs as building blocks. Many methods have been developed for the fabrication of CNTs based aerogels, including chemical vapor deposition (CVD) methods,[28,60] freeze-drying,[61,62] and critical-point-drying.[63–65] For example, Mateusz *et al.* fabricated a multiwalled carbon nanotube (MWCNT) aerogel by using a solution-based approach and steped by freeze-drying.[87] **Fig. 4** shows the fabrication procedures of the MWCNT aerogel. Pristine MWCNTs were first dispersed in chloroform by poly(3-hexylthiophene)-b-poly (3-(trimethoxysilyl) propyl methacrylate) aqueous solution by sonication. After that, the MWCNTs were crosslinked by polymers to form a wet gel. After the freeze-drying process, the wet gel was transformed into MWCNT aerogel. The obtained MWCNT aerogel exhibited a low mass density of 4 mg·cm^{-3}, a high electrical conductivity of 3.2×10^{-2} S·cm^{-1} and very large surface area of 580 m^2·g^{-1}. Meanwhile, the MWCNTs aerogel also showed an excellent mechanical properties with small thickness reduction after 1000 cycles of compression-recovery test. Recently, the fabrication of CNTs based hybird/composite aerogels are also studied extensively for a variety of applications. Qi *et al.* prepared a cellulose-CNTs hybrid aerogels by freezing-drying of the wet-gel precursors.[66] The composite aerogels showed excellent mechanical properties, thermal stability and good sensitivity to ambient pressure. They demonstrated that the cellulose-CNTs hybird aerogels can be used as sensors for the detection of gases or volatile organic compounds. Sun *et al.* fabricated an ultralight CNTs-graphene composite aerogels by directly freeze-drying of CNTs and graphene mixed aqueous suspension.[67] The 3D networks of the CNTs-graphene composite aerogels were constructed with giant graphene walls

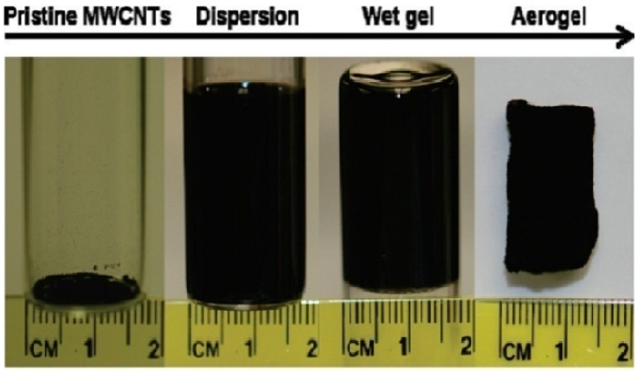

Fig. 4 Photographs shows the fabrication steps of MWCNTs aerogels.

Table 2 Characteristics of various CNTs aerogels based supercapacitors.

method	modification	surface area and pore size	capacitance	testing conditions	electrolyte	Ref.
CVD	microfibrous carbon	1059 $m^2·g^{-1}$ 1.41 nm	524 $F·g^{-1}$	- -	5 M KOH	60
CVD	-	90-100 $m^2·g^{-1}$ 3-3.4 nm	28.5 $F·g^{-1}$	1 $mV·s^{-1}$ three-electrode	1 M $LiPF_6$ in ionic liquid	69
CVD	PEDOT	- -	40 $F·cm^{-3}$	100 $V·s^{-1}$ three-electrode	1 M H_2SO_4	70
CVD	PANI	370 $m^2·g^{-1}$ 80 nm	189 $F·g^{-1}$	0.5 $A·g^{-1}$ two-electrode	1 M H_2SO_4	71
supercritical drying	cellulose nanofibers	871 $m^2·g^{-1}$ 1.3-50 nm	178 $F·g^{-1}$	5 $mV·s^{-1}$ two-electrode	H_2SO_4-PVA gel	72
freeze-drying	mesoporous carbon	557 $m^2·g^{-1}$ 6.6 nm	214 $F·g^{-1}$	10 $mV·s^{-1}$ three-electrode	1 M KOH	73
CVD	Ni-microfiber	155 $m^2·g^{-1}$ 3.6 nm	348 $F·g^{-1}$	0.2 $A·g^{-1}$ three-electrode	5 M KOH	74
CVD	carbon aerogel	670-710 $m^2·g^{-1}$ -	524 $F·g^{-1}$	1 mA three-electrode	5 M KOH	75

and CNTs ribs. The composite aerogels showed excellent integrated properties, such as outstanding elasticity, ultralow density ($\rho \geq 0.16$ $mg·cm^{-3}$), excellent thermal stability, good electrical conductivity, and high adsorption capacities toward organic compounds. The CNTs based aerogels not only hold the advantages of the CNTs, such as high electrical conductivities, high mechanical resilience and high thermal conductivity, but also exhibit the unique properties of aerogels (3D porous structure, low mass densities, continuous porosities, high surface areas). These features suggest that CNTs aerogels are suitable materials for the application as polarizable electrodes in supercapacitors. On the other hand, the 3D interconnected networks with a large number of porosity in the aerogels could provide a substrate for incorporating with other electrode materials such as activated carbon, metal oxide and conducting polymer, which further improves the capacitive properties of the aerogels.

In the past decade, there has been mushrooming growth of research focusing on the fabrication of novel CNTs based supercapacitors. The application of carbon nanotubes (CNTs) as electrode materials for supercapacitors was first reported by Niu *et al.* in 1997.[68] They demonstrated that nitric acid pretreated CNTs electrode exhibits a high surface area of 430 $m^2·g^{-1}$ and enhanced spesific capacitance of 102 $F·g^{-1}$. Recently, CNTs aerogels are studied as advanced electrode materials for supercapacitors due to their unique structure and properties. Li *et al.* fabricated a CNTs sponge (or aerogel) by using a CVD method.[69] The 3D CNTs sponge, consisting of interconnected conducting networks, can be easily compressed and recover most of its original scale (with small plastic deformation less than 5%). Cyclic voltammetry tests of the CNTs sponge based electrodes under compressive strain of 50% showed the specific capacitance maintained over 90% of the original capacitance, and more than 70% of the capacitance retained under 80% strain. The results demonstrated that the above compressible and deformation tolerant electrodes can be used in flexible supercapacitors. CNTs aerogels decorated with conducting polymers are also studied as electrode materials for supercapacitors. For example, Lee *et al.* fabricated a transparent and flexible hybird nanomembrane by coating CNT aerogel sheets with poly(3,4-ethylenedioxythiophene).[70] The hybrid nanomembrane exhibited very high mechanical strength and flexibility, and studied as electrode material for supercapacitors. The obtained supercapacitors show very large volumetric capacitance of ~40 $F·cm^{-3}$ at 100 $V·s^{-1}$, high volumetric energy of ~70 $mWh·cm^{-3}$ and power density of ~7910 $Wcm·cm^{-3}$. Polyaniline deposite on CNTs sponge networks were prepared by Zhong *et al.*[71] The free-standing and flexible polyaniline/CNTs sponge composites are applied as electrode material and deliver an excellent area capacitance of 1.85–1.62 $F·cm^{-2}$ with good cycle stability and high rate capability. CNTs based compositeaerogels prepared by incorporating CNTs with other carbonaceous materials such as cellulose nanofibers[72] and mesoporous carbon[73] are also fabricated and applied as electrode materials for supercapacitors. Deposition of CNTs arrays on other 3D networks by using a CVD method can also lead to the formation of CNTs based composite aerogels.[60,74,75] For instance, Fang *et al.* reported the synthesis of Ni-microfiber-supported CNT aerogel by a catalytic CVD method.[74] The CNT arrays are directly deposited on the surface of 3D Ni-microfiber networks. The obtained Ni-microfiber-supported CNT aerogel was further coated by polyimide and subsequently carbonized by pyrolysis to form a free-standing electreode. The composite electrode materials exhibited a high capacitance of 348 $F·g^{-1}$ at a current density of 0.2 $A·g^{-1}$ in 5 M KOH electrolyte. The high capacitance could be attributed to the unique 3D structure of the electrode and the doping with Ni. As an advanced material, CNTs based aerogels show many advantages such as robust mechanical properties, excellent flexibility, high conductivity and high surface area. More importanly, other materials with high capacitive performance can be easily incorporated into the networks of the aerogels to form composite materials. And these advantages make the CNTs based aerogels great promising as lightweight and flexible supercapacitors for real-world applications. In spite of these excellent properties, the high cost of production of the CNTs based aerogels still restricts their applications. The studies of CNTs aerogels or CNTs composite aerogels based supercapacitors are summarized in **Table 2**, where the synthetic methods and the capacitance are also listed.

3.3 Graphene based hydrogels and aerogels for supercapacitors

Graphene, a one-atom-thick 2D carbon material, has been studied extensively in various fileds due to its unique properties such as high mobility of charge carriers, excellent flexibility and intensity and superior thermal conductivity. The intrinsic capacitance of graphene was reported to be 21 mF·cm^{-2}. More importantly, graphene holds the advantages of large theoretical specific surface area (2630 m^2·g^{-1}), high conductivity and excellent chemical stability which provide a great potential for capacitance applications. For example, Zhu *et al.* prepared an activated graphene oxide (GO) with an extreamly high surface area of up to 3100 m^2·g^{-1} and a high electrical conductivity.[76] The activated GO was tested as electrode materials for supercapacitors and achieving a high specific capacitance of 166 F·g^{-1} and a high energy density of ~70 Wh·kg^{-1} in inonic liquid electrolyte. However, these unique properties of graphene are usually relevant at the nanoscale so that this restrains its macroscopic application. Especially, graphene nanosheets are easily agglomerate and restack into large paper-like structures which dramatically reduce its surface area and further harness its aplications in electrochemical electrodes and composite materials.

Gaphene based hydrogels and aerogels are 3D carbonaceous materials that prepared by using graphene sheets as building blocks. The graphene hydrogels and aerogels not only inherit the advantages of graphene but also hold the unique proeprties of the hydrogels and aerogels. Some strategies have been developed for the synthesis of graphene, including exfoliation from graphite, epitaxial growth and CVD. The epitaxial growth and CVD methods are difficult to manufacture graphene at a large scale and associated with problem of high cost. While the exfoliation method is suitable for scalable and low-cost production of graphene. Thus, exfoliated graphene has been widely used as building blocks for fabrication of graphene based 3D materials.

3.3.1 Graphene based hydrogels for supercapacitors. In recent years, gaphene based hydrogels and aerogels have been subject of intensive investigation for the application in supercapacitors. Generally, graphene based hydrogels are fabricated by using GO as building blocks. There are several methods for the gelation of GO, including self-assemble by hydrothermal reaction,[77] ions or polymers induced gelation[78] and cross-linking by chemical bonds.[79] For example, graphene hydrogels prepared by hydrothermal treatment of GO suspension are reported by Xu *et al.* (**Fig. 5**).[77] The graphene hydrogels exhibit a strong mechanical property, an excellent electric conductivity and a high spesific capacitance of about 175 F·g^{-1}. Similarly, other researchers also reported the self-assembled graphene hydrogels prepared by hydrothermal reaction, exhibiting high spesific capacitance at range of 186~308 F·g^{-1}.[80–84] Graphene hydrogels can also be fabricated under mild conditions by using small molecules as reducing agent. For instance, self-assembled graphene hydrogels have been successfully fabricated by chemical reduction of GO with L-glutathione[85] and sodium ascorbate,[86] respectively. An asymmetric supercapacitor based on the L-glutathione reduced graphene hydrogels and nanostructured MnO$_2$ showed a specific capacitance of 157.7 F·g^{-1} at current density of 1 A·g^{-1}. While, the sodium ascorbate reduced graphene hydrogels based electrode exhibited a specific capacitance of 240 F·g^{-1} at current density of 1.2 A·g^{-1}. Recently, Yang *et al.* fabricated a densily packed graphene hydrogel films by using a capillary compression method.[87] They confirmed that the flexible graphene hydrogel films hold advantages of low ion transport resistance and high ion-accessible surface area, leading to excellent performance for supercapacitors (255.5 F·cm^{-3} in aqueous electrolyte and 261.3 F·cm^{-3} in organic electrolyte at current density of 0.1 A·g^{-1}). The high performance of the densily packed graphene hydrogel films hold great potential for large-scale real-world applications due to the fact that the methods are essentially compatible with the traditional paper-making process and can be readily scaled up.

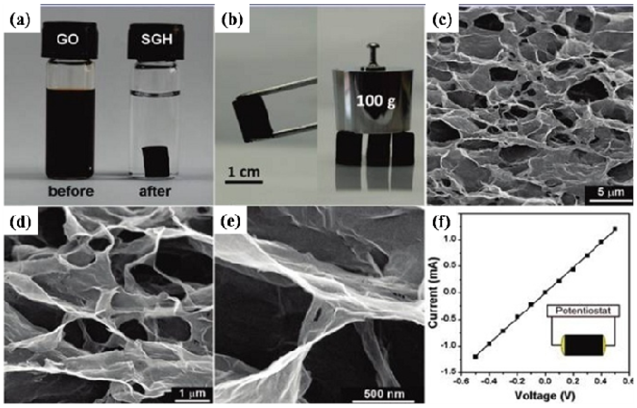

Fig. 5 (a) Photograph shows the GO susupension before and after hydrothermal reaction. (b) Digital pictrues show the graphene hydrogels are easily lifted and loaded with weight. (c–e) SEM images of the networks of the graphene hydrogel. (f) *I–V* curve of the graphene hydrogel.

The porous structure of graphene hydrogels could allow the decoration of another nanomaterial into the 3D networks to form graphene hydrogels based composite materials. To further improve their applicability, composites, consisting of graphene hydrogels with other materials such as conducting polymer,[88–90] metal oxide,[91] and CNTs,[92] could be expected to possess high performance for supercapacitors due to the synergetic effects. For example, Zhou *et al.* fabricated a hybrid hydrogel containg graphene and poly(3,4-ethylenedioxy-thiophene) (PEDOT).[89] They demonstrated that the hybrid hydrogel exhibit an excellent mechanical performance, a high specific capacitance of 174.4 F·g^{-1} and a good electrical conductivity of 0.73 S·cm^{-1}. These predominant properties could be due to the synergetic combination of the excellent conducting, mechanical properties of graphene and the pseudo-capacitance of PEDOT. Surface functionalization of graphene hydrogels with molecules that can undergo reversible redox reactions could also lead to an enhanced capacitance for supercapacitors. Xu *et al.* prepared a modified graphene hydrogel by using hydroquinone as functionalizing molecules, achieving an impressive specific capacitance of 441 F·g^{-1} at 1 A·g^{-1}.[93] Recently, graphene/Ni(OH)$_2$ composite hydrogels has been successfully fabricated and applied as 3D electrode materials for

Table 3 Characteristics of various graphene hydrogels based supercapacitors.

method	modification	surface area and pore size	capacitance	testing conditions	electrolyte	Ref.
hydrothermal	-	- -	175 $F\cdot g^{-1}$	1 $A\cdot g^{-1}$ two-electrode	5 M KOH	77
chemical reaction	ethylene diamine	745 $m^2\cdot g^{-1}$ 47 nm	232 $F\cdot g^{-1}$	1 $A\cdot g^{-1}$ two-electrode	2 M KOH	79
hydrothermal	organic amine	- -	190.1 $F\cdot g^{-1}$	10 $A\cdot g^{-1}$ two-electrode	5 M KOH	80
hydrothermal	urea	>1300 $m^2\cdot g^{-1}$ 1.7-4.3 nm	308 $F\cdot g^{-1}$	3 $A\cdot g^{-1}$ three-electrode	6 M KOH	81
hydrothermal	hydroxylamine	- -	205 $F\cdot g^{-1}$	1 $mV\cdot s^{-1}$ two-electrode	25% KOH	82
hydrothermal	-	414 $m^2\cdot g^{-1}$ 2-70 nm	186 $F\cdot g^{-1}$	1 $A\cdot g^{-1}$ two-electrode	H_2SO_4-PVA gel	83
hydrothermal	-	951 $m^2\cdot g^{-1}$ -	220 $F\cdot g^{-1}$	1 $A\cdot g^{-1}$ two-electrode	5 M KOH	84
chemical reduction	L-glutathione	315.2 $m^2\cdot g^{-1}$ 2-10 nm	157.7 $F\cdot g^{-1}$	1 $A\cdot g^{-1}$ two-electrode	0.5 M Na_2SO_4	85
chemical reduction	sodium ascorbate	- -	240 $F\cdot g^{-1}$	1.2 $A\cdot g^{-1}$ three-electrode	1 M H_2SO_4	86
filtration	-	- -	255.5 $F\cdot cm^{-3}$	0.1 $A\cdot g^{-1}$ two-electrode	1 M H_2SO_4	87
chemical reduction	PANI	- -	334 $F\cdot g^{-1}$	2 $A\cdot g^{-1}$ three-electrode	6 M KOH	88
polymer cross-linking	PEDOT	- -	104 $F\cdot g^{-1}$	0.5 $A\cdot g^{-1}$ two-electrode	1 M Na_2SO_4	89
hydrothermal	PPy	463 $m^2\cdot g^{-1}$ -	330 $F\cdot g^{-1}$	1.5 $A\cdot g^{-1}$ three-electrode	3 M $NaClO_4$	90
hydrothermal	Co_3O_4	- -	757.5 $F\cdot g^{-1}$	0.5 $A\cdot g^{-1}$ three-electrode	6 M KOH	91
hydrothermal	CNTs	237 $m^2\cdot g^{-1}$ -	318 $F\cdot g^{-1}$	0.1 $A\cdot g^{-1}$ two-electrode	30% KOH	92
hydrothermal	hydroquinones	297 $m^2\cdot g^{-1}$ 2-70 nm	441 $F\cdot g^{-1}$	1 $A\cdot g^{-1}$ two-electrode	1 M H_2SO_4	93
hydrothermal	$Ni(OH)_2$	92 $m^2\cdot g^{-1}$ -	1247 $F\cdot g^{-1}$	5 $mV\cdot s^{-1}$ three-electrode	6 M KOH	94
deposition	nickel foam	1260-1725 $m^2\cdot g^{-1}$ -	45.6 $mF\cdot cm^{-2}$	0.67 $mA\cdot cm^{-2}$ two-electrode	5 M KOH	95

supercapacitors.[94] This hybrid hydrogels exhibited a high specific capacitance of ~1247 $F\cdot g^{-1}$ at a scan rate of 5 $mV\cdot s^{-1}$ with excellent cycling stability. Deposition of graphene hydrogels on 3D networks of nickel foams are also applied as composite electrodes for supercapacitors.[95] The composite electrode materials show a high capacitive performance which could be due to synergetic effects of the high conductivity and electrochemical stability of graphene hydrogel and the short distance of charge transfer from graphene hydrogel to the current collecting electrodes. The studies of graphene hydrogels based supercapacitors are summarized in **Table 3**, where the preparing method and the capacitance are given.

3.3.2 Graphene based aerogels for supercapacitors. Replacing the infilling medium, water, of graphene hydrogel with air without collapsing the network structure can lead to the 3D networks, graphene aerogel. Generally, graphene aerogels are prepared by supercritical or freeze drying of the graphene based wet gels. For instance, Chen *et al.* fabricated a reduced-GO (rGO) aerogels by gelation of the rGO sheets in aqueous solution and stepped by freeze-drying.[96] The obtained rGO aerogels exhibit a 3D porous structure with high mechanical strength. The study of graphene aerogels for supercapacitors has been booming in recent years due to the fact that graphene aerogels are large 3D networks with continuous porous structure and large surface area. The unique structure of graphene hydrogels could provide efficient ionic transportation and adsorption, leading to high performance for supercapacitors.

Recently, rGO aerogels have been successfully prepared and studied as electrode materials for supercapacitors. The rGO aerogels can be obtained by thermal reduction[97] or H_2 reduction[98] of a GO aerogel, or by pre-reduction of GO in the wet gels stepped by freeze-drying or supercritical drying. Chemicals such as mercaptoacetic,[96] L-ascorbic acid[99] and hypophosphorous acid/I_2[100] are applied for the gelation and reduction of GO. Liu *et al.* fabricated a flexible and folded structured graphene paper from the thermal reduced GO

method	modification	surface area and pore size	capacitance	testing conditions	electrolyte	Ref.
freeze-drying	-	- -	172 $F\cdot g^{-1}$	1 $A\cdot g^{-1}$ two-electrode	1 M H_2SO_4	97
supercritical-drying	-	870 $m^2\cdot g^{-1}$ 2-50 nm	153 $F\cdot g^{-1}$	0.1 $A\cdot g^{-1}$ three-electrode	Ionic liquid	98
freeze-drying	L-ascorbic acid	512 $m^2\cdot g^{-1}$ 1.5-55 nm	128 $F\cdot g^{-1}$	0.05 $A\cdot g^{-1}$ two-electrode	6 M KOH	99
supercritical-drying	hypophosphorous acid	830 $m^2\cdot g^{-1}$ 4 nm	278.6 $F\cdot g^{-1}$	0.2 $A\cdot g^{-1}$ three-lectrode	1 M H_2SO_4	100
freeze-drying	nitrogen and boron co-doping	249 $m^2\cdot g^{-1}$ -	62 $F\cdot g^{-1}$	5 $mV\cdot s^{-1}$ three-electrode	H_2SO_4-PVA	101
freeze-drying	carbohydrates	12.5-364.6 $m^2\cdot g^{-1}$ -	161.6 $F\cdot g^{-1}$	0.5 $A\cdot g^{-1}$ three-lectrode	Na_2SO_4	102
freeze-drying	metal oxide	350 $m^2\cdot g^{-1}$ 2-3.5 nm	226 $F\cdot g^{-1}$	1 $mV\cdot s^{-1}$ three-lectrode	1 M H_2SO_4	103
pyrolization	carbon	361-763 $m^2\cdot g^{-1}$ 10-50 nm	122 $F\cdot g^{-1}$	0.05 $A\cdot g^{-1}$ two-electrode	6 M KOH	104

aerogels.[97] They demonstrated that the graphene paper based electrodes showed better capacitive performance compared to available carbon materials. Moreover, these kind of electrodes are more flexible, free-standing, binder-free, and suitable for mass production.

Many researchers focus on the fabrication of garphene aerogels based composite materials for supercapacitors. For example, Wu *el al.* synthesized a nitrogen and boron co-doped graphene aerogels by introducing ammonia boron trifluoride in the wet gels and stepped by freeze-drying.[101] They fabricated an all-solid-state supercapacitor by using the doped graphene aerogels as electrode materials and PVA/H_2SO_4 gel as electolyte. The all-solid-state supercapacitors showed a specific capacitance of about 62 $F\cdot g^{-1}$, an energy density of about 1600 $W\cdot kg^{-1}$ and an excellent cycling stability. Organic moleculers or polymers are also applied for the modification of graphene aerogels. For instance, carbohydrates, including glucose, β-cyclodextrin and chitosan are employed for the synthesis of hybrid graphene-based aerogels.[102] The carbohy-drates modified graphene aerogels showed an excellent electrochemical behavior with strong and stable performance. Metal oxides are also introduced into the networks of graphene aerogels to form composite materials. Wu *et al.* fabricated graphene based composite aerogels by decorating of metal oxides such as Co_3O_4 and RuO_2 in the networks.[103] The metal oxides loaded graphene aerogels exhibited an outstanding specific capacitance of 226 $F\cdot g^{-1}$, a high rate capability and an excellent cycling stability, due to the synergetic effects of the pseudo-capacitive metal oxides and high surface area, high conductivity and electrochemical stability of the graphene aerogels. Meng and co-workers prepared graphene/carbon composite aerogels by imbedding graphene into the precursor of carbon aerogels (resorcinol-formaldehyde) and stepped by pyrolyzing.[104] The composite aerogels exhibited a desirable specific capacitance (122 $F\cdot g^{-1}$ at current density of 50 $mA\cdot g^{-1}$) with excellent rate performance. The reports of graphene aerogels based supercapacitors are summarized in **Table 4**, where the synthetic methods and the capacitance are shown.

3.4 Biomass-derived hydrogels and aerogels for supercapacitors

Biomass derived carbonaceous materials have attracted significant interests due to its low cost, easily accessible and environmental friendly. Biomass derived hydrogels and aerogels are a large class of gels consisting of 3D solid networks and a lot of open pores. Benefiting from their unique structure and excellent properties, biomass derived gels have been studied extensively in supercapacitors both as electrolyte and electrode materials. Generally, the biomass-derived hydrogels can be used as gel electrolytes and as binders for fabrication of composite electrode materials. While the biomass derived aeogels are usually applied as electrode materials for supercapacitors. Biomass derived carbonaceous hydrogels can be prepared by hydrothermal treatment of biomass[6,105] or directly obtained from the crude natural polymers.[106] As for binders' application, the 3D hydrogels are served as substrate for dispersion of electrochemical active materials to form a composite electrodes. For example, DNA hydrogels were utilized as template for the binding of electrode materials,[107] and

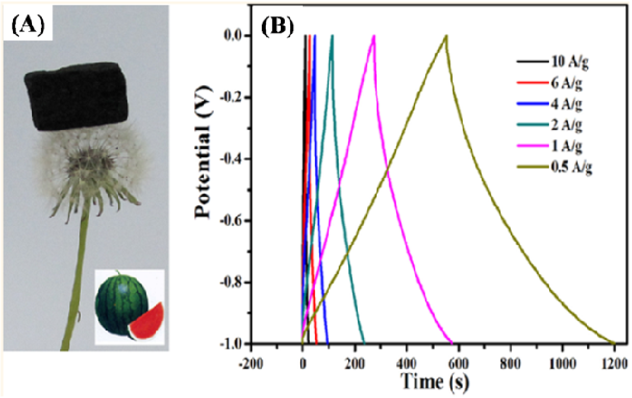

Fig. 6 (a) Photograph shows the ultralight magnetic carbon aerogel that can be supported by a dandelion. (b) Charge-discharge curves of the magnetic carbon aerogel based electrodel at different current densities.

chitosan hydrogels binding black pearls carbon were employed as electrodes for fabrication of all solid-state electrical double layer capacitors.[106] Owing to the synergetic combination of the biocompatability, the unique structure and strong mechanical properties, the biomass derived hydrogels could have great potential in supercapacitor for the application as power source for implantable biomedical devices. The biomass derived carbon-aceous hydrogels are also widely used as electrolyte materials due to the fact that the 3D solid polymeric networks can adsorb and retain water. For example, Choudhury *et al.* reported the application of a natural gel, gelatin hydrogel, as solid electrolytes for supercapacitors.[108] The supercapacitor displayed an optimal capacitance of 81 $F{\cdot}g^{-1}$. Yamazaki and co-workers fabricated novel composite hydrogels containing two kinds of natural materials, cellulose and chitin, and two kinds of ionic liquids.[109] The hybrid hydrogels immersed with H_2SO_4 aqueous solution were applied as electrolytes for supercapacitors. The gel electrolyte based supercapacitors displayed an enhanced capacitance of 155 $F{\cdot}g^{-1}$ with excellent high-rate discharge capability and durability. The advantages of good relability, good biocompatibility and absence of leakage of liquids could endow them great potentials in supercapacitors for biomedical applications.

Biomass derived carbonaceous hydrogels and aerogels are also studied extensively as electrode materials for supercapacitors due to the synergetic benefits obtained from combination of the low-cost, sustainable biomass and the unique structure, large surface area and strong mechanical proeprties of the hydrogels and aerogels. Recently, we have fabricated carbonaceous gels by using crude biomass, watermelon as the source.[6] The obtained carbonaceous hydrogels and aerogels showed an interconected microscopic structure with average pore diameter of about 45.8 nm. We further decorated Fe_3O_4 nanoparticles into the 3D network of the aerogels and transformed it into magnetic carbon aerogels by pyrolyzing. The magnetic carbon aerogels showed a very light weight and exhibited an excellent capacitance of 333.1 $F{\cdot}g^{-1}$ and an outstanding cycling stability for supercapacitors (**Fig. 6**). As a natural hydrogel, bacterial cellulose has attracted an increasing interests as new materials or substrates for a wide range of applications. The study of bacterial cellulose (BC) based electrode materials are also boomed in recent years. For example, Lee *et al.* fabricated a nano carbons by using the BC as carbon source.[110] The nano carbons based electrodes displayed a specific capacitance of 42 $F{\cdot}g^{-1}$ and an area-normalized specific capacitance of 1617 $F{\cdot}cm^{-2}$. Other BC based composite materials are also studied as electrodes for supercapacitors, such as BC/PANI nanocomposites[111] and BC/CNTs composite papers.[112] Cellulose nanofibers are also studied extensively for the fabrication of composite aerogels. For example, a hybrid aerogels containing cellulose nanofibers and rGO are fabricated by Gao et al.[22] The obtained cellulose nanofibers/rGO hybrid aerogels are applied as electrode materials for supercapacitors, achieving a high capacitance of 207 $F{\cdot}g^{-1}$. Similarly, Cellulose nanofibers–multi-walled CNTs aerogels are also synthesized and showed a specific capacitance of about 178 $F{\cdot}g^{-1}$.[113]

4 Summary and outlook

In this feature article, we provide a comprehensive review of the recent advance of the applications of carbonaceous hydrogels and aerogels for supercapacitors. The fabrication of all kinds of carbonaceous hydrogels and aerogels, including polymer derived carbonaceous gels, carbon nanotubes based carbonaceous gels, graphene based carbonaceous gels and biomass derived carbonaceous gels are discussed and the related synthetic methods are summarized. The application of the carbonaceous hydrogels and aerogels in supercapacitors are introduced and their performance are listed and compared. The polymer derived carbon aerogels have the advantages of high specific surface area and easily tuning the pores which are benificial to achieve a high performance for supercapacitors. The rich in resource is another advantage for the CAs. However, the poor mechanical properties hinder them for the applications in flexible supercapacitors. CNTs based aerogels and graphene based aerogels hold advantages of high conductivity, robust mechanical properties, excellent flexibility and high surface area. More importantly, many of the aerogels are free-standing, binder-free and ease for mass production. These advantages make them great promising as electrodes for lightweight and flexible supercapacitors. These supercapacitors are suitable power source for portable and wearable electronic devices. In spite of these excellent properties, the high cost of raw materials and difficultt for production still restrict their applications. Biomass derived carbonaceous gels hold the advantages of abundant in resources, low price and environmental friendly. However, the limited energy storage and rate capability restrain them for the applications in high performance supercapacitors.

As we can seen, the unique structure and excellent properties of the carbonaceous gels could provide great application potential for supercapacitors. However, how to enhance their performance for supercapacitors is still a hot topic and the fabrication of composite electrode materials with more efficiency and sustainability is still challenge. More importantly, urgent efforts should be made for the fabrication of flexible and wearable supercapacitors to meet an increasing demand of the market for portable and wearable electronic devices such as sportswear, wearable displays and embedded health monitoring. Significant progress in the development of novel type energy storage systems have been achieved over the past decade. As one of the promising energy storage systems, supercapacitors have achieved great success for the applications as power source with high-performance and low-cost. Thanks to the rapid expansion of nanoscience, many neotype electrochemical active materials have been developed, especially the novel carbonaceous nanomaterials. As the development and flourishing of these novel carbon materials (such as CNTs and graphene) which provide great opportunities for the development of new energy storage systems or devices. Carbonaceous hydrogels and aerogels as a large class of porous nanocarbons would also play an important role in the fabrication of neotype energy storage devices. Thus, we believe the applications of carbonaceous hydrogels and aerogels for next-generation energy storage systems such as supercapacitors, will meet its golden age.

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TOC:

This feature article gives an overview of the recent advances of using carbonaceous hydrogels and aerogels for supercapacitors.