An Overview of Stretchable Supercapacitors Based on Carbon Nanotube and Graphene

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Abstract The wearable demand of modern electronic devices makes flexible and stretchable energy storage device urgently needed. Stretchable and flexible supercapacitors (SCs) are energy storage devices that provide ultrahigh power density while having long-term durability, high security, and electrochemical stability. Among different SCs electrode materials, CNTs and graphene-based materials exhibit great potential in terms of stretchable SCs due to its ultrahigh electrical conductivity, large specific surface area and good mechanical properties. In this review, the state-of-the-art process and achievements in the field of stretchable SCs enabled by CNTs and graphene are presented, which include the novel design strategy, mechanical and electrochemical properties. The final section highlights current challenges and future perspectives on research in this thriving field.

Keywords Stretchable; Supercapacitors; Carbon materials; Graphene; CNTs

INTRODUCTION

In the past decade, with the rapid development of intelligent electronic devices, people demand more convenient and smarter wearable electronic devices, like smart watch, flexible electronic screen and Google project glass.\textsuperscript{[11–17]} As an important part of flexible electronic equipment, achieving flexibility and extensibility of energy storage device is an urgent problem. However, it is difficult to make conventional secondary batteries flexible and stretchable due to its security issues.\textsuperscript{[8,9]} Supercapacitors (SCs), as one of the promising energy storage equipment, is featured with higher power density and super long durability, which can provide high power supply for wearable electronic equipment due to its reliable safety, controllable size and harmless to the human body.\textsuperscript{[10–13]}

According to the energy storage mechanism and the type of electrode materials, SCs can be classified into three types: electrical double-layer capacitors (EDLCs),\textsuperscript{[14,15]} pseudocapacitors (PCs),\textsuperscript{[16,17]} and hybrid capacitors.\textsuperscript{[18,19]} The energy storage mechanism of EDLCs is an electrostatic process in which the electrostatic charge accumulates at the interface between the electrode surface and electrolyte. Generally, porous carbon nanomaterial such as graphene,\textsuperscript{[20–24]} carbon nanotube,\textsuperscript{[25–29]} activated carbon,\textsuperscript{[30–33]} mesoporous carbon,\textsuperscript{[34–38]} and carbide-derived carbon,\textsuperscript{[39,40]} have been widely used in EDLCs. The specific surface area of the electrode material is the key factor to determine the capacity, which ensures higher powder density, fast charge-discharge rate, higher rate capability and excellent long cycling stabilities. However, the limited specific surface area and porous distribution of carbon nanomaterial often cause a relatively low energy density. On the other hand, the pseudocapacitors present a chemical process with fast and reversible redox reactions occurring on the surface of the electrode material, which can offer higher energy density.\textsuperscript{[41]} The composite materials composed of carbon nanomaterials and conducting polymers (e.g., polypyrrole (PPy),\textsuperscript{[42–45]} polyaniline (PANI),\textsuperscript{[46–49]} and poly(3,4-ethylenedioxythiophene) (PEDOT)\textsuperscript{[50–53]} or transition metal oxide (e.g., RuO\textsubscript{2},\textsuperscript{[54–56]} MnO\textsubscript{2},\textsuperscript{[57–59]} Co\textsubscript{3}O\textsubscript{4},\textsuperscript{[60–62]} NiO,\textsuperscript{[63–65]} TiO\textsubscript{2},\textsuperscript{[66–68]} and VO\textsubscript{2}\textsuperscript{[69–72]} ) have been widely implied for the electrode. However, the structural instability and poor conductivity during the electrochemical reaction of such pseudocapacitor materials will lead to poor rate capability and cycling life. Moreover, the intrinsic mechanical performance of transition metal oxide makes them not suitable for flexible and stretchable supercapacitors.

Carbon nanotube (CNT) and graphene are sp\textsuperscript{2}-boned carbon and considered as the basic construction material for carbon materials. The surface-dangling bonds such as the COOH, OH, or C\textequiv O group enable CNT and graphene to be ideal candidates for the stretchable SCs. Owing to its unique structural
As a typical carbon nanomaterial, CNTs have been widely investigated as a promising electrode material. CNTs have a cylindrical structure with a nanometer-scale diameter, which can be categorized as single-walled nanotubes (SWCNTs) and multi-wall nanotubes (MWCNTs). Based on its unique one-dimensional (1D) structure, the CNTs have superior physical properties, such as a high charge mobility of $1.0 \times 10^6$ cm$^2$V$^{-1}$s$^{-1}$, thermal conductivity of 3500 Wm$^{-1}$K$^{-1}$, Young’s modulus of 950 GPa and theoretical specific surface area of 1315 m$^2$g$^{-1}$. And CNTs can be assembled into macroscopic CNT architectures with different dimensions, including 1D fibers, 2D films, and 3D foams. In addition, these macroscopic CNT materials are very suitable for ideal electrode material for high-performance stretchable SCs based on its adjustable specific surface area and abundant micro-nano structure. Moreover, many efficient methods have been established for the mass preparation of CNTs recently, including chemical vapor deposition (CVD), arc discharge and laser ablation. This enables CNTs to be produced in a larger-scale at relatively low cost and widely used in academic and industrial field.

As the most basic type of structure, 1D CNT nanofibers can be easily handled and utilized into a stretchable SCs. And they can be woven into cloth, which endows them with special applications in wearable electronics. Recently, a lot of interesting work for designing wire-shape CNTs has been done to expand the capability of elastic fiber based stretchable SCs. Xu et al. designed a stretchable SC consisting of two wavy-shaped CNT fibers twisted together on a PDMS spandex fiber. Fig. 1(a) depicts the structure of a wire-shaped SC (WSS). The “prestraining-then-buckling” fabrication process (Fig. 1b) is used to prepare the stretchable WSS. First, a thin layer of PDMS was coated on the spandex fiber, which formed a core-skin structure. The inner spandex fiber provided the elasticity and the outer PDMS layer protected the inner fiber from the gel electrolyte. Second, the core-skin composite fiber was prestretched to 100%, and a thin layer of $\text{H}_2\text{SO}_4$-PVA gel electrolyte was applied onto the PDMS/spandex fiber. Finally, a straight WSS was “glued” onto the prestrained PDMS/spandex fiber. The stretchable SC exhibited an area specific capacitance of 4.63–4.99 mFcm$^{-2}$, and the ability to undergo large cyclic tensile strain of 100%. Coaxial structure CNTs fiber is another way to achieve stretchable SCs. Yang et al. studied a fiber based stretchable SC by wrapping aligned CNT sheets on an elastic fiber (Fig. 1c). The fiber-shaped supercapacitor maintained a high specific capacitance of approximately 18 Fg$^{-1}$ after stretching by 75% for cycles.

The electrochemical performance of stretchable SCs used CNTs as electrode material is simply far from meeting the requirements of practical application. The introduction of pseudocapacitive electrode material is an effective method for improving the performance of CNTs-based stretchable SCs. Chen et al. coated a thin layer of PEDOT/poly(styrene sulfonate) (PEDOT-PS) onto the CNTs-wrapped elastic wires, which exhibited an extremely high elasticity of up to 350% strain with an improved capacitance of 30.7 Fg$^{-1}$. In addition, Choi et al. used electrochemical deposition method for fabricating the MnO$_2$ nanofiber-doped CNTs, which were wrapped on the nylon sewing thread. The composite carbon nanotube/MnO$_2$/polymer fiber solid-state SC exhibited a length, area and volume-normalized specific capacitance of 5.4 mFcm$^{-1}$, 40.9 mFcm$^{-2}$, and 3.8 Fcm$^{-3}$, respectively. However, there are several problems for the stretchable SCs. For example, the electrode material should have a high capacitance, low resistance and enough volume for the intercalation of electrolyte. In order to solve these problems, many strategies have been proposed.
SCs with noncapacitive elastomeric polymers as stretchable matrix. The whole volume and weight of the device are high, which is not suitable for application in portable and wearable electronic; the elastomeric polymers also have poor mechanical properties and low operating temperature. Given all the components, the device exhibits relatively low capacitance and energy density. Thus, construction of self-stretchable, lightweight SCs for modern electronic device with elastic polymer substrates is critical and challenging. Zhang et al. reported fiber springs as freestanding elastic SCs and lithium-ion batteries with high electrochemical performance.[80] As shown in the Fig. 2(a), the spring-like fiber electrode comprised twisted aligned multiwalled CNTs with uniform coiled loops, which provided the electrode with high stretchability and elongation of over 300%. The stretchable SC was fabricated by placing two springlike fibers in parallel, which could be bent in any direction without degradation in performance. Additionally, Shang et al. fabricated a substrate-free, highly flexible SC based on two twisted CNT yarns consisting of uniformly arranged micro-loops. This helical yarn structure was also self-stretchable and exhibited a large strain (up to 150%).

Besides, the novel CNT yarns showed a good dynamic property by stretching the CNT yarns to large strain under low to high frequency (0.2−10 Hz).[81] In some special application scenarios, the functionalized fiber-shaped stretchable SCs have also attracted great attention. With the development of self-healing stretchable electrolyte, the self-healing and stretchability come into being. Huang et al. developed an electrolyte comprising poly(acrylic acid) dually crosslinked by hydrogen bonding and vinyl hybrid silica nanoparticles.[82] This electrolyte displayed all superior functions and provided a solution to the intrinsic self-healability and high stretchability problems of a SC. Benefiting from the intrinsic stretchability, self-healability and ionic conductivity of the new polyelectrolyte, supercapacitors based on this polyelectrolyte possessed intrinsic self-healability while the capacitance was completely maintained during all 20 breaking/healing cycles (Fig. 3a). A great variety of yarns with different mechanical and electrical properties can be found in the textile industry, potentially resulting in the development of a diverse array of yarn-based SCs. The wearable and wovenable urethane plastic fiber core spun yarns (UY),

![Figure 2](https://example.com/fig2.png)

**Fig. 2** (a) SEM image of a spring-like fiber as different magnifications and a fiber at different strains of 0%, 50%, and 100% (Reprinted with permission from Ref. [80]; Copyright (2014) John Wiley and Sons). (b) Fabrication of self-stretchable fiber-shaped SCs and SEM images of a 6.9 mm-long supercapacitor in original state ($l_1 = 6.9$ mm, $\varepsilon = 0$) and after stretched to $l_2 = 13.6$ mm, $\varepsilon = 100\%$, showing uniform deformation of helical loops, and a continuous electrolyte coating on the yarns surface (Reprinted with permission from Ref. [81]; Copyright (2015) Elsevier).

![Figure 3](https://example.com/fig3.png)

**Fig. 3** (a) Fabrication of the patch-assisted non-autonomic self-healable supercapacitor (Reprinted with permission from Ref. [82]; Copyright (2015) Nature). (b) Digital photographs of the urethane yarn (UY) and common cotton yarn-based fabrics at unstretched (inserted photos) and stretched states (Reprinted with permission from Ref. [83]; Copyright (2016) Elsevier).
formed by entwining native cotton yarns around highly elastic urethane filaments, were introduced to offer intrinsic high stretchability and act as a wearable substrate for hosting conductive CNTs and electrocapacitive poly pyrrole (PPy).[83] When the yarns were woven into a fabric, the UY-based SC can be stretched to 300% strain while the common cotton yarn-based can only be reached by 20% (shown in Fig. 3b).

Based on the excellent mechanical properties of CNTs, the CNTs film can be easily fabricated, which can be used to fabricate the plane-shaped stretchable SC. The “prestrain-stick-release” is the most common strategy. As shown in the Fig. 4(a), a stretchable macrofilm SC was fabricated by three steps: (1) prestretching of an elastomeric substrate of a PDMS; (2) exposure to UV light to form a hydrophilic surface; (3) releasing of the prestrain in the PDMS substrate for spontaneous formation of periodically buckled patterns. The formed wrinkled structure (Figs. 4b and 4c) made the SCs highly stretchable without sacrificing electrochemical performance.[84] The initial specific capacitances were 54 F·g⁻¹ for the stretchable SCs without applied strain and 52 F·g⁻¹ for that subject to 30% applied strain. Remarkably, the specific capacitances of the stretchable SCs did not change under 1000 charge-discharge cycles, which exhibited excellent electrochemical stability.[84] This is because the buckling wavy film could effectively enhance the interaction with electrolyte and inhibit the decrease of active specific area. In order to enhance the specific capacitance and the working potential, designing CNT-based hybrid electrodes has proven to be an effective method. Gu et al. reported MnO₂/CNT dynamically stretchable SC with increased specific capacitance of 100–119 F·g⁻¹ as the scan rate increased from 50 mV·s⁻¹ to 200 mV·s⁻¹.[85] Meanwhile, the electrochemical performance of MnO₂/CNT stretchable SC was stable under the dynamic mechanical stretching-releasing (DSR) processes at high strain rate of 6% strain per second. However, strain rate slightly affected the electrochemical performance, with a small decrease of charge transfer resistance (Rct) from 0.15% strain per second to 6% strain per second, indicating an improved electrode/electrolyte interface at a relatively high strain rate.[85]

However, such symmetric stretchable SCs still suffer from low specific capacitance and low working potentials, leading to unsatisfactory power and energy densities. Building of asymmetric electrode configuration has been demonstrated as an effective method for improving the electrochemical performance of stretchable SC. It takes advantage of the pseudocapacitance positive electrode to improve the specific capacitance as well as the CNTs negative electrodes to broaden the operating potential. Tang et al. fabricated an asymmetric stretchable SC based on wrinkled CNT@MnO₂ cathode and CNT@poly pyrrole anode with electrochemical performance. The asymmetric device displayed remarkably enhanced energy density of 40.0 Wh·kg⁻¹ at the power density of 519 kW·kg⁻¹ with the strain of 100% and a signification enhancing the potential window of 0–2.0 V.[86] Besides, a stretchable all-solid-state asymmetric SC based on wrinkled
MnO$_2$/CNT and Fe$_3$O$_4$/CNT macrofilms was fabricated by Gu et al. As shown in Fig. 5(a), the MnO$_2$/CNT film, Fe$_3$O$_4$/CNT film, and Na$_2$SO$_4$/PVA gel electrolyte were used as the positive electrode, negative electrode, and electrolyte, respectively. MnO$_2$/CNT electrode exhibited a potential window of 0−1.2 V, while that of the Fe$_3$O$_4$/CNT electrode was in the range of −0.8–0 V (as shown in Fig. 5b). Thus, the potential window of the asymmetric SC could be extended to 2.0 V (Fig. 2c). In addition, the stretchable asymmetric SC showed an excellent cycling stability with only 1.1% deterioration of the initial specific capacitance after 1.0 × 10$^4$ cycles and a mechanical stability (shown in the Figs. 5d and 5e).

The stretchable SCs fabricated by depositing the active material onto the prestretched substrate cannot achieve a high maximal stretched strain because a high recovery stress may destroy the deposited materials. Therefore, how to regulate the composition and structure of materials to improve the electrochemical and mechanical performance of the stretchable SCs remains a challenge. The assembly of CNT array as stretchable SCs is an effective strategy to solve this problem. Peng et al. reported a nitrogen-doped core-sheath CNT array for highly stretchable SC. As shown in Fig. 6(a), a CNT array was first synthesized from ethylene at 740 °C by chemical vapor deposition (CVD). The CNTs array was regrown coaxially with N-doped layers by CVD again but at a higher temperature of 1060 °C. Then, polyurethane (PU) was coated on the N-doped CNTs array forming an NCNT/PU film, followed by deposition of PVA gel electrolyte. As the elastic polymer, PU can easily infiltrate into NCNTs, which is important for realizing a high stretchability (Fig. 6b). The SCs exhibited a high specific capacitance of 31.1 mF·cm$^{-2}$. It is worth mentioning that the specific capacitance was maintained by 98.9% after stretching to a strain of 400% and 96% after stretching for 1000 cycles at a strain of 200%. The stretchable SCs mentioned above can only be stretched in one direction. However, the stretchable SCs retaining high electrochemical performance during multi-directional stretching are essential for many applications. Cao et al. reported a crumpled-CNT forest stretchable SCs with good electrochemical performance and stability under either uniaxial (300%) or biaxial strains (300% × 300%) for thousands of stretching-releasing cycles. Fig. 6(c) shows the fabrication of crumpled-CNT forest electrode. The growth of CNT forest on silicon wafer is similar to the previous works. The elastomer substrate used in this work is either uniaxially or biaxially prestretched, which is the key factor to fabricate uniaxially or biaxially (two orthogonal in-plane directions) stretchable SCs. The stretchable SCs with an elastic substrate displays mechanical mismatch between the active electrode material and the substrate during stretching. In addition, the substrate adds extra non-capacitive weight and volume to the device, which would decrease the energy density. The design of CNT-based film with specific structure can achieve stretchable SCs without elastic substrate. As shown in Fig. 7(a), He et al. fabricated a stretchable SC based on CNT-film with cellular structure. The specific capacitance of the flexible and freestanding SCs could be maintained by 98.3% for 3000 cycles under 140% strain. It was demonstrated that the novel SCs...
could be used as power supply for various wearable and portable device. As shown in Fig. 7(b), the stretchable SCs were able to serve as a “watch strap” for a commercial electronic watch. After charging, the “watch strap” could drive the electronic watch. Additionally, due to high stretchability, this device was able to work at different sizes of wrists (Fig. 7b).

Through the same method, the researchers fabricated another type of three-dimensionally stretchable SCs by designing a novel pyramid structure. Compared to the previous stretchable SCs that could only be stretched in plane, this freestanding pyramid-structured SCs could be stretched in three dimensions for thousands of cycles.

**GRAPHENE-BASED STRETCHABLE SCs**

Graphene is considered as the basic construction material for carbon materials. Owing to its unique structural features (a one-atom-thick 2D single layer of sp²-bonded carbon), graphene...
possesses a series of prominent intrinsic chemical and physical characteristics, such as extremely high electron mobility (2.0 × 10^6 cm^2·V^−1·s^−1),[93] strong mechanical strength (~1 TPa), and large specific surface area (2675 m^2·g^−1).[95] Based on intriguing and outstanding features, graphene-based materials exhibit huge potential for high-performance portable and stretchable SCs.

A variety of graphene films based on different preparation methods have been demonstrated that can be used as high-rate and long cycle stability stretchable SCs due to its excellent flexibility and resistance to tensile. Zang et al. reported a simple and time-saving method for fabricating extremely stretchable and high-performance SCs based on crumpled-graphene papers, which can be stretched biaxially.[98] As the substrate was prestretched, the crumpled graphene paper unfolded (Figs. 8a and 8b), which benefits the mechanical reliability of graphene paper under multiple cycles of large deformation. The crumpled graphene paper based SCs showed a unique combination of high stretchability (linear strain ~300%, areal strain 800%), high specific capacitance (196 F·g^−1) and high reliability (1000 stretch/relax cycles).

Compared to the CNT-based film, due to the exceptional flexibility and transparency, chemical vapor deposition (CVD) growing graphene films have been regarded as an ideal candidate for stretchable transparent electrodes. Chen et al. reported a transparent and stretchable SC based on wrinkled graphene electrode, which showed a high transparency (57% at 500 nm) and could be stretched up to 40% without any capacitance attenuation over hundreds of stretching cycles.[96] In order to improve the transparency of transparent electrode, Xu et al. fabricated ultrathin CVD-grown graphene film for stretchable, transparent and high-rate SCs.[97] The transparent and stretchable device based on bucked four-layer graphene film realized the unique combination of good stretchability (up to 40%), high optical transparency (72.9% transmittance), and excellent electrochemical performance with high rate capability and cycle stability (98% retention: 98% after 10 × 10^4 cycles). Designing wavy-shaped graphene films is helpful to improve the specific area between graphene electrode and electrolyte, which leads to enhanced electrochemical properties of graphene-based stretchable SCs. Xie et al. fabricated a wavy shaped polyaniline/graphene electrode used Ni foam as a catalyst.[98] This stretchable SC exhibited a maximum specific capacitance of 261 F·g^−1, which is higher than the one based on a flat shaped or crumpled shaped graphene films. Besides, Qi et al. reported suspended wavy graphene microribbons, which overcome the limitation that micro supercapacitors are stiff but only the interconnection conductor is stretchable.[99] The specific capacitance of the resulting micro supercapacitors remained almost unchanged when stretched under 100%, even after 5000 stretching/releasing cycles. This outstanding performance is mainly due to two designs: (1) the suspended wavy structured electrode arrays relieved the stain concentration on the electrode fingers in the stretching/relaxing process, which prevented the happening of detachment and crack; (2) distance was kept constant in the stretching/relaxing process, which further enhanced the stability.[99]

However, the complex routes and high cost involved in the fabrication of graphene limit its commercialization prospect as graphene-based energy storage device. Recently, a novel laser-induced technique at room temperature has been applied to fabricate graphene-based stretchable SCs. Lamberti et al. reported a laser direct writing technique to prepare laser-induce graphene.[100] This material consisted of a 3D network of multilayer graphene in the laser writing process, during which the sp^2-carbon atoms in the polymer were photothermally converted to sp^3-carbon atoms. The resulting stretchable SCs showed good mechanical performance in the stretching and bending process, from 0% to 50% and from 0° to 160°, respectively. In addition, this device showed good cycling stability, retaining 84% of its original capacitance after 1000 cycles in stretching condition and almost 90% in the bending condition. Moreover, stretchable micro-supercapacitors (MSCs) can greatly facilitate the development of modern smart electronic devices due to their facility to package and well-suited integration with microelectronic systems.[101,102] A highly stretchable and transparent all-solid-state MSC was fabricated by the active material on silicone rubber substrates, which consisted of 3D laser-induced graphene network and conductive PEDOT. The device exhibited an enhanced specific capacitance of 790 µF·cm^−2 at the discharge current of 50 µA·cm^−2.[103]

Fiber shaped materials play a critical role in our lives and have been widely used as supercapacitor electrodes with many advantages of being wearable into lightweight, highly flexible, soft, and low-cost textiles. Flexible graphene fibers stand for a new type of fiber, which integrates many unique

![Fig. 8](https://doi.org/10.1007/s10118-020-2386-x)
properties such as high strength, electrical and thermal conductivities of individual graphene sheets into the useful, macroscopic ensembles. However, it remains a challenge for the assembling of 2D graphene sheets with irregular size and shape into macroscopic fibrillar configuration.\(^{104–106}\) As a result, relatively few studies have been reported on the stretchable fibrous graphene SCs. Recently, Meng et al. reported a facile one-step strategy to fabricate graphene fiber within glass pipeline.\(^{107}\) The graphene fiber had a low density of 0.23 g·cm\(^{-3}\), which is 7 times and 85 times lower than that of the conventional carbon fiber and Au wire, while remaining strong, flexible, conductive, weavable, and their unique structure with multifunctionalities can be further modified in an in situ or post-synthesis fashion. The graphene fiber was further decorated with 3D graphene as a sheath through electrolyzing GO aqueous suspension (as illustrated in Fig. 9a). Due to the highly exposed surface areas, high electrical conductivity, and good chemical stability, the specific capacitance of spring SC based on core-sheath all graphene fiber changed little under 50% compression and 200% stretch.\(^{107}\) Recently, graphene-woven fabric fabricated by CVD method was used as a dynamically stretchable solid-state SC. This stretchable SC based on graphene-woven fabric can be stretched up to 30%·s\(^{-1}\) dynamically with excellent galvanic stability, which still kept stable even under 60%·s\(^{-1}\) strain rates.\(^{108}\) However, the reliability and the stability of SC were generally destroyed by mechanical damage and deformation in practical application. Developing self-healability stretchable SC is extremely urgent. Wang et al. reported reduced graphene fiber-based springs wrapped with self-healing polymer outer shell as a stretchable and self-healing SC, which is illustrated in Fig. 9(b). The SC kept 82.4% capacitance after 100% stretch and 54.2% capacitance after third healing.\(^{109}\) The hydrophobicity and high stretchability of stretchable SCs are also important in the practical application. Guo et al. reported a highly stretchable waterproof fiber asymmetric SCs with an integrated structure.\(^{110}\) This fiber SC demonstrated a working voltage of 1.6 V and a high energy density of 2.86 mW·cm\(^{-3}\). This excellent electrochemical performance remained unchanged after the SC was immersed in water for 50 h, and retained 95% of its initial capacitance after 3000 cycles of stretching-releasing at a maximum strain of 400%. Graphene oxide is an ideal framework to assemble metal, small molecules, and polymer due to the multiple functional groups on its surface, such as phenol hydroxyl, epoxide groups, and carboxylic acid. The fabrication strategy of the stretchable SC mentioned above is simply mixing the graphene and stretchable polymer, involving no formation of chemical bond between graphene and stretchable polymer. Very recently, Li et al. fabricated a macromolecularly interconnected 3D graphene/nanostructured conductive hydrogels.\(^{111}\) The possible interactions between the polymer and graphene oxide involved electrostatic interaction, hydrogen bond and π-π stacking (as illustrated in Fig. 9c). The graphene/nanostructured conductive polymer SC showed a remarkable volumetric energy density of 8.8 mWh·cm\(^{-3}\), which outperforms many fiber-shaped SCs in the references. And this proof-of-concept all hydrogel design opens up opportunities in the field of next-generation stretchable and wearable SCs. In the end, the electrochemical and stretchable performances of stretchable SCs with different types are sum-

Fig. 9 (a) Schematic illustration of a wire-shaped SC fabricated from two twined GF@3DGs with polyelectrolyte (Reprinted with permission from Ref. [107]; Copyright (2014) Nature). (b) Schematic diagrams of the stretchable and self-healable mechanism, manufacturing process of PPy/RGO/MWCNT electrodes, and stretchable and self-healing supercapacitor (Reprinted with permission from Ref. [109]; Copyright (2016) American Chemical Society). (c) The possible interaction among PANI, PA, and RGO (Reprinted with permission from Ref. [111]; Copyright (2019) John Wiley and Sons).

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| Electrode          | Substrate | Electrolyte          | Specific capacitance (F·g⁻¹) | Areal capacitance (μF·cm⁻²) | Energy density (mWh·cm⁻³) | Power density (mA·cm⁻²) | Working potential (V) | Cycle life                  | Reference |
|-------------------|-----------|----------------------|-----------------------------|-----------------------------|----------------------------|-------------------------|------------------------|--------------------------|-----------|
| MCNT              | Spandex   | H₂SO₄                | 4.63−4.99                   | 0.8 × 10⁻⁴−2.26 × 10⁻⁴      | 4.93 × 10⁻¹                 | 0−0.8                  | 100% (1.0 × 10⁴ cycles at 100% strain) | [76]       |
| MCNT              | Elastic fibers | H₃PO₄/PVA gel | 19.2 (at 0.1 A·g⁻¹)           | 0.363−0.515                         | 19−421                     | 0−0.8                 | 95% (1000 cycles at 75% strain) | [77]       |
| SWCNT             | PDMS      | Polymer              | 55.3 (at 0.5 V·s⁻¹)           | 4.2                          | 0.5                        | 0−3                     | 100% (at 30% strain) | [112]       |
| SWCNT             | PDMS      | H₂SO₄/PVA gel        | 82                           | 0.5 mF·cm⁻³                    | 3.7 mWh·cm⁻³               | 6200 mW·cm⁻³            | −0.8−0.8                | 100% (1000 cycles at 50% strain) | [113]       |
| Aligned CNT fiber and sheet | Nylon | PVA-LiCl gel | 59                           | 8.66                         | 1.88 Wh·kg⁻¹              | 755.9 W·kg⁻¹            | 0−1                    | 81.6% (75 cycles at 10% strain) | [114]       |
| CNT               | −         | H₃PO₄/PVA gel        | −                            | 40.9 (at 10 mV·s⁻¹)           | 2.6                       | 6.69 × 10⁴              | 0−1                    | 150% strains of 6% (12%, and 1.7% strain per second) | [79]       |
| MnO₂/CNT hybrid fiber | KOH-PVA | −                     | 27.07 (at 150 mA·cm⁻²)        | −                           | −                         | 0−1                    | 94% (300 cycles at 100% strain) | [80]       |
| CNT/Ppy           | −         | Gel                  | 63.6 (at 100 mV·s⁻¹)          | −                           | −                         | 0−1                    | 99% (1.0 × 10⁴ cycles at 100% strain) | [115]       |
| CNT/Ppy           | −         | Polymer              | 85 (at 500 mV·s⁻¹)            | −                           | −                         | 0−0.6                  | 220% in a cycle of 600% strain | [82]       |
| CNT/Ppy           | −         | Urethane             | Na₂SO₄                      | 69                           | 6.13 × 10⁻³                | 1.33                    | 0−0.8                  | 85% (1000 cycles at 40% strain) | [83]       |
| CNT/PAETOT-PPS/MnO₂ | PDMS     | H₃PO₄/PVA hydrogel  | 30.7 (at 0.5 A·g⁻¹)          | −                           | −                         | 0−0.8                  | 105% (100 cycles at 200% strain) | [78]       |
| CNT/PAETOT-PPS/MnO₂ | PDMS     | Gel                  | −                           | 7                           | 33.5 × 10⁻³                | 0−0.8                  | 100% (1000 cycles at 30% strain) | [116]       |
| MONT/MnO₂         | −         | LiCl-PVA gel         | 278.6                       | 125.37                      | 0−0.8                    | 0−0.8                  | 92% (3000 cycles at 100% strain) | [117]       |
| SWNT/MnO₂/Ppy     | −         | Gel                  | 461                         | 31.1 Wh·kg⁻¹                 | 22.1 kW·kg⁻¹              | 0−0.8                  | 96.2% (7.5 × 10⁴ bending cycles) | [119]       |
| Buckled MnO₂/CNT  | PDMS      | TEABF₄/PC            | 92                          | −                           | −                         | −1.5−1.5               | 99.33% (5000 cycles at 6% strain) | [85]       |
| CNT/Ppy           | −         | CH₂=CH-SiO₂/PAAM     | 637 (at 1 mA·cm⁻¹)           | 40 Wh·kg⁻¹                  | 519 kW·kg⁻¹              | 0−2                    | 96% (500 cycles at 100% strain) | [86]       |
| MnO₂/CNT Fe₂O₃/CNT | PDMS     | Na₂SO₄/PVA gel      | 130.2                       | 45.8 Wh·kg⁻¹                 | 0.41 kW·kg⁻¹              | 0−2                    | 98.9% (2000 cycles at 0% strain, 6000 cycles at 100% strain, 2000 cycles at 50% strain) | [87]       |
| CNT/PAETOT-PPS/MnO₂ | PDMS     | H₃PO₄/PVA gel       | 42.4                        | 72.9 (at 0.5 mA·cm⁻¹)        | 0.701 mWh·cm⁻³           | 337.0 mW·cm⁻³           | 0−1                    | 98.3% (3000 cycles at 140% strain) | [91]       |
| CNT/Graphene      | −         | H₃PO₄/PVA gel       | 152.4                       | 2900                         | −                         | −1                    | 91% (under 100% uniaxial strain)88% (under 100% biaxial strain) | [120]       |
| Electrode Substrate | Electrode | Electrolyte | Specific capacitance (F·g⁻¹) | Areal capacitance (mF·cm⁻²) | Energy density (mWh·cm⁻³) | Power density (mW·cm⁻²) | Working potential (V) | Cycle life | Reference |
|---------------------|-----------|-------------|--------------------------------|-----------------------------|----------------------------|------------------------|----------------------|------------|-----------|
| SWCNT/TiO₂          | PDMS      | PVA/LiCl gel| 15.1 (at 1 V·s⁻¹)             | 2.16                        | 0.05                       | 0–0.8                  | 100% (100 cycles at 200% strain) | [121]     |
| CNT alloy           | PU        | H₂PO₄/PVA gel| 31.1                          | 2.11                        | 0.54                       | 0–0.8                  | 100% (100 cycles at 200% strain) | [88]       |
| CNT/MoS₂            | PDMS      | H₂PO₄/PVA gel| 10.67                         | 13.16 F·cm⁻¹                | 0.59                       | 0–0.8                  | 100% (500 cycles at 100% strain) | [90]       |
| CNT/AgS₃            | PDMS      | H₂PO₄/PVA gel| 85.3                          | 64                          | 0.83                       | 0–0.8                  | 100% (500 cycles at 100% strain) | [122]     |
| PEDOT/doped CNT      | PDMS      | H₂PO₄/PVA gel| 5 (at 50 mV·s⁻¹)              | 5.58                        | 36.3                       | 0–0.8                  | 100% (500 cycles at 100% strain) | [89]       |
| VHB 4910            | PVA/KCl gel| 261.5       | 36.3                          | 5.58                        | 36.3                       | 0–0.8                  | 100% (500 cycles at 100% strain) | [123]     |
| Graphene/PEO/PANI   | PDMS      | H₂PO₄/PVA gel| 121                          | 26                          | 5.33                       | 0–0.8                  | 97% (4000 cycles with different e at 1.0 and 1.3) | [126]     |
| RGO/PANI            | Ni        | H₂PO₄/PVA gel| 261                          | 3.8 Wh·kg⁻¹                 | 804 W·kg⁻¹                 | 0–0.8                  | 95% (100 cycles at 30% strain) | [98]       |
| Graphene micofiber  | H₂SO₄/PVA gel| 40             | 1.7 × 10⁻⁴                    | 0.1                         | 0–0.8                     | 100% (500 bending cycles (bending radius of ~2 mm)) | [107]     |
| Graphene woven fabric | PDMS | H₂PO₄/PVA gel| 17.3                         | 5.33                        | 2.8 × 10⁻³                  | 36.48                   | 0–0.8                  | 95% (100 cycles at 30% strain) | [95]       |
| Graphene oxide/MCNT/PPy | PDMS | H₂PO₄/PVA gel| 7.6                          | 5.8                         | 0.54                       | 0–0.8                  | 95% (100 cycles at 30% strain) | [107]     |
| Graphene microribbons | PDMS | H₂PO₄/PVA gel| 196                          | 3.65                        | 0−0.8                      | 84% (1000 cycles at 50% strain) | [100]     |
| Graphene paper      | PDMS      | NaCl/PVP gel| 7.6                          | 5.8                         | 0−0.8                      | 84% (1000 cycles at 50% strain) | [100]     |
| Graphene            | PDMS      | PAAK/KOH     | 7.6                          | 5.8                         | 0−0.8                      | 84% (1000 cycles at 50% strain) | [100]     |
| N-Graphene polymer  | PDMS      | Gel          | 0.54 (at 500 mV·s⁻¹)         | 0.52                        | 41.7                       | 0–0.8                  | 92% (5000 cycles at 50% strain) | [99]       |
| Graphene/MoS₂       | PDMS      | PVA/LiCl gel| 567                          | 720 F·cm⁻¹                  | 77.8 Wh·kg⁻¹               | 0–2                    | 91.2% (100 cycles at 100% strain) | [128]     |
| Graphene/PANI       | PDMS      | H₂PO₄/PVA gel| 70                          | 1.728 mWh·cm⁻³               | 0.062 W·cm⁻³               | 0–0.8                  | 87% (300 cycles at 30% strain) | [127]     |
| rGOF/MnO₂           | PVA/LiCl gel| 40.7          | 3.52                         | 0–1.6                      | 91.2% (100 cycles at 100% strain) | [128]     |
| rGOF/PANI hydrogel  | H₂SO₄/PVA gel| 193.65       | 8.8 mWh·cm⁻³                 | 30.77 W·cm⁻³               | 0–0.8                    | 95% (3000 cycles at 400% strain) | [110]     |
marized in Table 1. Stretchable capacitors based on carbon nanotubes and graphene exhibit good electrochemical properties and mechanical stability, but this is far from practical application in the modern life. It is an efficient method for improving the electrochemical performances of stretchable SCs by incorporating carbon nanomaterials with metal oxides or conductive polymer.

CONCLUSIONS AND OUTLOOK

In this review, the recent advances and process about the fabrication and electrochemical property of CNT and graphene-based stretchable SCs were summarized. In general, the stretchable capacitors currently reported are mainly based on CNT and graphene-based fiber or film and the stretchable polymer substrates. Furthermore, it is an effective method to improve the electrochemical performance of the stretchable SCs through introducing pseudocapacitance materials, such as conductive polymers (PEDOT, PANI, PPy) and metal oxide (MnO₂, NiO, Co₃O₄, and so on). Based on their good electrical conductivity, interlinked structure and unique elasticity modulus, CNT-based stretchable SCs exhibit good electrochemical properties while also having good mechanical tensile properties. Compared to CNT, graphene as the stretchable SCs electrode material has a higher specific surface area, which can offer more electrochemical reaction sites and enhance the contact area with electrolyte. At the same time, the abundant functional groups on graphene oxide enable it to be easily combined with conductive polymers and metal oxide, which can further enhance the electrochemical performance of the whole graphene-based stretchable SCs. CNT and graphene still play important roles in the research of next-generation stretchable SCs due to their excellent electrochemical performance and superb mechanical robustness. In addition, the development of stretchable SCs without elastic substrate is one of the important directions in the future, which is beneficial to the energy density due to its reduced weight of the whole stretchable SCs device. Meanwhile, the stretchable SCs without polymer would enhance environmental friendliness and weather fastness. For example, based on 3D printing technology, a highly stretchable neat carbon aerogel compromised of graphene and CNT with a retractable 200% elongation was reported by Gao et al. which is expected to be used in stretchable SCs. The integration of electronic equipment leads to the miniaturization of capacitor equipment. Hence, flexible and stretchable micro-SCs based on silk-screen printing, 3D printing technology and other advanced technologies will be the future trend. With the continuous process of novel technology, the wearable electronic devices based on carbon nanomaterials will play an important role in our future lives.

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