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Abstract
The development of wearable devices such as smart watches, intelligent garments, and wearable health-monitoring devices calls for suitable energy storage devices which have matching mechanical properties and can provide sufficient power for a reasonable duration. Stretchable fiber-based supercapacitors are emerging as a promising candidates for this purpose because they are lightweight, flexible, have high energy and power density, and the potential for easy integration into traditional textile processes. An important characteristic that is oftentimes ignored is stretchability-fiber supercapacitors should be able to accommodate large elongation during use, endure a range of bending motions, and then revert to its original form without compromising electrical and electrochemical performance. This article summarizes the current research progress on stretchable fiber-based supercapacitors and discusses the existing challenges on material preparation and fiber-based device fabrication. This article aims to help researchers in the field to better understand the challenges related to material design and fabrication approaches of fiber-based supercapacitors, and to provide insights and guidelines toward their wearability.

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Elastic Fiber Supercapacitors for Wearable Energy Storage

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Abstract

The development of wearable devices such as smart watches, intelligent garments and wearable health monitoring devices calls for suitable energy storage device which has matching mechanical properties and provide sufficient power for a reasonable duration. Stretchable fiber-based supercapacitor is emerging as a promising candidate for the purpose due to its lightweight, flexibility, high energy and power density, and easy integration into traditional textile processes. In addition, an ideal stretchable fiber supercapacitor should be able to accommodate large elongation during use, endure a range of bending motions, and then revert to its original form without compromising the electrical and electrochemical performance. This article summarizes the current research progress on stretchable fiber-based and discusses the existing challenges on material preparation and fiber-based device fabrication. This article aims to help researchers in better understanding the challenges related to stretchable fiber-based supercapacitors, and provide insights and guidelines towards truly wearable electronic devices.

1. Introduction

Wearable and portable electronic products represent a new and important field of modern electronics. These products are demonstrated in a variety of applications and devices, which include smart watches, flexible displays, intelligent garments and wearable health monitoring devices.[1-4] As a result of these exciting advancements, the development of suitable energy storage devices including supercapacitors,[5-8] batteries[9, 10] and solar cells[11, 12] are becoming increasingly necessary to power these electronics. Among these power sources, supercapacitors (SCs) are preferred over others when such characteristics as high power density, fast charging-discharging rate, good cycling stability, and low maintenance are critical.[8, 13, 14] However, conventional SCs are typically rigid and heavy representing a mismatch in the basic requirements of wearable and portable devices such as flexibility and lightweight. It is therefore critical to develop new types of SCs that are flexible, lightweight and have the energy storage performance comparable or better than that of the current technology.

1.1 The emergence of flexible fiber-shaped supercapacitors

The development of flexible SCs such as thin film SCs for flexible electronic devices have emerged as a fruitful research area. Flexible film SCs were initially derived from conventional SCs and possess a number of structural similarities. By reducing the thickness of the device and replacing the rigid current collector with flexible substrates, many flexible fiber SCs prototypes have been developed and their integration into flexible devices have been demonstrated.[15-17] Recent advances in wearable electronics and energy storage devices have resulted in the emergence of novel fiber-based SCs.

Fiber-shaped SCs usually appear in a shape similar to a one-dimensional wire with diameters ranging from a few micrometers to millimeters. These fiber-based SCs are

lightweight and thus provide high aerial, volumetric and gravimetric energy storage performance. Some of the fiber-based SCs make use of active materials, like graphene and carbon nanotubes, transition metal oxides and dichalcogenides, conducting polymers, and many other types of two-dimensional layered nanomaterials have reasonable degrees of flexibility *i.e.* they can be repeatedly bent without significantly altering their energy storage performance.[15, 16, 18-22] Some of these fiber-based SCs have been hand-woven in textile prototypes, albeit in short lengths, but sufficient to demonstrate textile integration.[14, 17, 21, 23-27] Although flexible, present fiber electrodes do not have enough stretchability and elasticity to meet the real demands of a true wearable technology. An ideal fiber SC should accommodate large elongation during use, endure a range of bending motions, and then revert back to its original form (elastic) without compromising the electrical and electrochemical performance.[28, 29] The mismatch in mechanical requirements originates from the inherent rigidity and brittleness of the active materials used in making the fiber electrodes.

Imparting stretchability and elasticity to the fiber electrodes is therefore one of the major challenges in the development of a truly functional fiber-based SCs. To date, the two general approaches in achieving stretchable/elastic fiber-based SCs involve integrating a stretchable/elastic material platform with the active materials or using structural engineering of the fiber electrodes. In the first approach, a typical practice is to deposit carbon nanotube (CNT) electrodes by wrapping onto elastic substrate like polyurethane rods or rubber/rubber-like rods.[24, 30-34] In the second approach, structural engineering is achieved by geometrical re-configuration of straight filaments into wire-like helical or coiled structures and this oftentimes involves twisting and coiling.[17, 23, 35-38] In some of these cases, other active materials such as MnO_2 and polyaniline are incorporated by suitable coating techniques to increase the overall electrochemical performance of the device.[31, 32, 35]

It is also apparent from recent developments that rod-shaped rubber-like SCs are not ideal platforms for textile-based wearable technologies because of their limited length, low scalability and particularly poor wearability. One stand-out elastic material candidate for textile integration is polyurethane-based (PU). PUs represents a family of elastomeric polymers that can be easily spun using industrially scalable wet-spinning technologies into continuous filaments. The commercially available Spandex fiber is one prominent example of PU with widespread use in clothing industry particularly in athletic sportswear, compression garments and medical fabrics.[39-41] This wide-spread use makes PU particularly attractive for the development of elastic fiber-based SCs. However, PU fibers are inherently electrically insulating. The research challenge is to make electrically conductive PU, and more importantly, to ensure that the added conductivity can also provide the means to store energy (*i.e.* as electrochemical supercapacitor). Recent literature shows that PU fibers can be made conductive by incorporating electrically conductive fillers such as metals nanowires (silver),[42] nanocarbon materials (carbon black, CNT, graphene)[43, 44] and conductive polymers (PEDOT:PSS).[45, 46] These novel conductive PU fibers have been produced by wet-spinning, and have been shown useful in applications as stretchable fiber conductors and strain sensor textiles.[46] However, the electrical conductivity of these novel fibers is still low to be used for applications in energy storage. In addition, the electrochemical properties of these fibers are still unexplored.

1.2 From flexible to stretchable fiber-based supercapacitors

In general, fiber-based SCs are inherently flexible due to their fiber-shaped structure and small diameter. However, common fiber-based SCs based on conductive fibers and carbon-based current collector cannot sustain large stretching deformation due to the rigidity and brittleness of the electrode materials (*e.g.* < 7 % for PEDOT:PSS fibers, < 3 % for graphene fibers, ~ 10 % for graphene/CNT fibers).[16, 17, 19] It was estimated that during normal physical

activity such as walking, skin on feet and waist stretch and contract by as much as 55 %. For certain types of application such as compression garment and body sensors, it is thus reasonable to expect that a stretchable fiber-based SC must endure repetitive bending and twisting, and sustain stretching deformation greater than 50 %. There is currently no clear definition and distinction of flexible and stretchable fiber-based SCs in literature. We recommend that fiber-based SCs that are highly flexible but can only sustain a stretching deformation of < 10 % to be categorized into flexible fiber-based SCs, while those able to sustain a stretching deformation of > 50 % and reverse back to the original length to be categorized as stretchable fiber-based SCs.

This article will discuss recent progress on stretchable and elastic fiber-based SCs. The discussions will cover the knowledge gaps, challenges and potential solutions on electrode materials, electrode fabrication and performance enhancements. The limitations in mechanical properties and stability of performance of current flexible fiber SCs for wearable applications will also be discussed. This article will also provide history and insights on the evolution of flexible and stretchable fiber-based SCs and opportunities in the field for application in wearable technologies. It is important to note that the scope of this article is on elastic/stretchable fiber-based SCs, for there are already a number of reviews that cover broader topics including flexible SCs and fiber-based SCs.[47-49] We hope that this article will help researchers in better addressing the challenges related to elastic and stretchable fiber-based SCs so that they can ultimately work towards truly wearable electronic devices.

2. Approaches for imparting elasticity and stretchability to fiber-based supercapacitors

The fiber conductivity is a prerequisite in making stretchable/elastic fiber SCs. Common elastic fibers such as PU fiber and rubber fiber are electrically insulating. Electrically conductive elastomeric fibers can be produced by coating or by blending electrically conductive materials such as metals nanowires (*e.g.* silver), nanocarbon materials (*e.g.* carbon black, CNT, graphene) and conducting polymers into elastomeric fibers.[42, 43, 45] For example, stretchable conductive fibers can be produced by coating a thin layer of conductive carbon black/natural rubber composite onto a PU fiber,[50] or by coating with a sheath of graphene/PVA composites. [51] These fibers can be stretched as much as 500 % and possess electrical resistance as low as 0.25 kΩ cm^{-1}.[51] Stretchable conductive fibers can also be fabricated by blending conductive materials into the elastomer matrix.[42] Highly stretchable conductive fibers from poly(styrene-β-isobutylene-β-styrene) (SIBS) as elastomer and poly(3-hexylthiophene) (P3HT) as conducting filler have been prepared by wet-spinning, which exhibited a high electrical conductivity of ~0.38 S cm^{-1} and sustained a maximum tensile strain of 975 %.[52] Seyedin *et al.* produced all-polymer stretchable conductive fibers by wet-spinning of composite formulations of PU and PEDOT:PSS which showed an electrical conductivity of 13 S cm^{-1} and an elongation break at of 259 %.[45] Successful incorporation of nanocarbon materials (graphene, CNT and carbon black) into PU fibers using wet-spinning had also been reported.[43, 44]

Apart from the electrical conductivity, stretchable fiber-based electrode should also have excellent energy storage capability in order to power electronic devices for a reasonable duration. To date, the two general approaches in achieving stretchable/elastic fiber-based SCs involve integrating stretchable/elastic material platforms with active materials or using

structural engineering to create elasticity and stretchability. There are also reports that employed a combination of both approaches in realizing the stretchability within fiber-based SCs. A summary and comparison of stretchable fiber-based SCs including their capacitance and stretchability are presented in **Table 1**. Some examples are discussed in the following section.

2.1 Elastic fibers coated with capacitive materials

Using elastic fibers as the building platform is probably the most facile route to produce stretchable fiber-based SCs. Coatings of conductive and electrochemical active materials on the fiber surface imparts the elastic fiber with capacitive property. This approach takes advantage of the readily available commercial elastic fibers and has other advantages including low cost and scalability.

Since elastic fibers only act as a stretchable substrate and are generally insulating, researchers have exploited different structures to build stretchable fiber-based SCs. For example, in a coaxial structure, two electrodes are positioned at different layers of the coating, and gel electrolyte is used to separate the two electrodes from short circuiting. Yang *et al.* designed a stretchable coaxial fiber SC with a rubber fiber as an elastic substrate and CNT sheets as electroactive materials.[34] The aligned CNT sheets were wrapped on the rubber fiber with a gel electrolyte (**Figure 1a-b**). The fiber SC demonstrated a specific capacitance of 20 F g^{-1}, an energy density of 0. 515 W h kg^{-1}, and a power density of 421 W kg^{-1}. The fiber SC can be easily stretched to 100 % without an obvious decrease in its structural integrity. The capacitance of the fiber SC was maintained at 18 F g^{-1} after 100 stretching cycles under an applied strain of 75 %. Many other coaxial stretchable fiber-based SCs have been developed based on similar methods. Stretchable fiber SC produced by wrapping pre-stretched elastic rubber fiber with PANI modified CNT sheets was reported.[32] The produced

fiber SC could be stretched by over 400 % (**Figure 1c**) and the specific capacitance of 79.4 F g^{-1} retention was 80 % after stretching at a strain of 300 % for 5000 cycles.

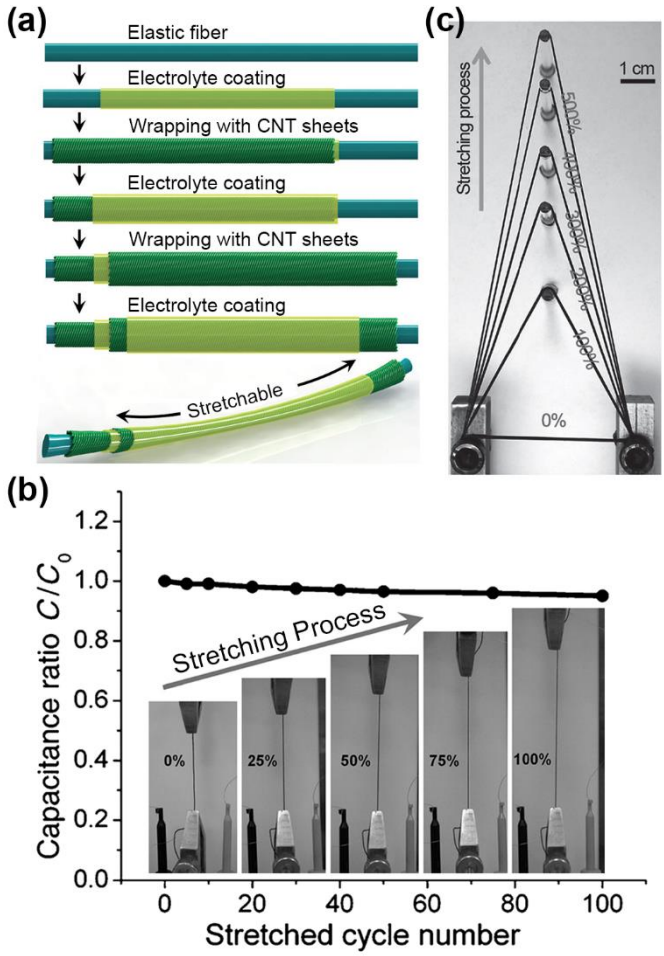

Figure 1. (a) Schematic illustration of the fabrication process of a highly stretchable fiber SC in a coaxial structure and (b) the relative capacitance (C/C_0) of the fiber SC as a function of stretching cycles at the strain of 75 % (insert shows photographs of fiber SC at tensile strains of 0, 25, 50, 75, 100 %).[34] Copyright © 2013 WILEY-VCH. (c) photograph of the fiber electrodes before and after stretching by 100 %, 200 %, 300 %, 400 % and 500 %.[32] Copyright © 2015 WILEY-VCH.

Researchers also have constructed fiber-based SCs with helical and parallel structures where two separated fibers act as the positive and negative electrodes respectively. Sun *et al.* explored the used of low-cost natural cotton fiber.[53] A stretchable fiber-based yarn SC was produced by co-spinning of elastic PU fiber and cotton fiber into a stretchable yarn, dip-coating of CNTs onto the yarn and subsequently electrodepositing polypyrrole (PPy) on the CNTs-coated yarn (**Figure 2a–b**). The yarn SC could deliver a high areal capacitance of 69

mF cm^{-2} and withstand a strain of 80 % without significant impact on the capacitance. It retained 85 % of its initial capacitance after 1000 stretching cycles with an applied strain of 40 %. This stretchable fiber SC showed much higher capacitance and improved durability during stretching cycles compared to the neat CNT fiber. However, its capacitance was still low for application and it could only sustain low strain during stretching cycles. Chen *et al.* reported a stretchable fiber SC consisting of two fiber electrodes in a helical structure.[33] Fiber electrodes were prepared by wrapping PEDOT:PSS coated CNT sheets on polyester-polyurethane fibers. A fiber-based SC was fabricated by twisting two fiber electrodes and coating with gel electrolyte (**Figure 2c-d**). The resulting stretchable fiber SC exhibited a high elasticity sustaining up to 350 % strain with a specific capacitance of 30.7 F g^{-1}. It also showed slightly enhanced capacitance (6 %) after 100 stretching cycles with an applied strain of 200 %.

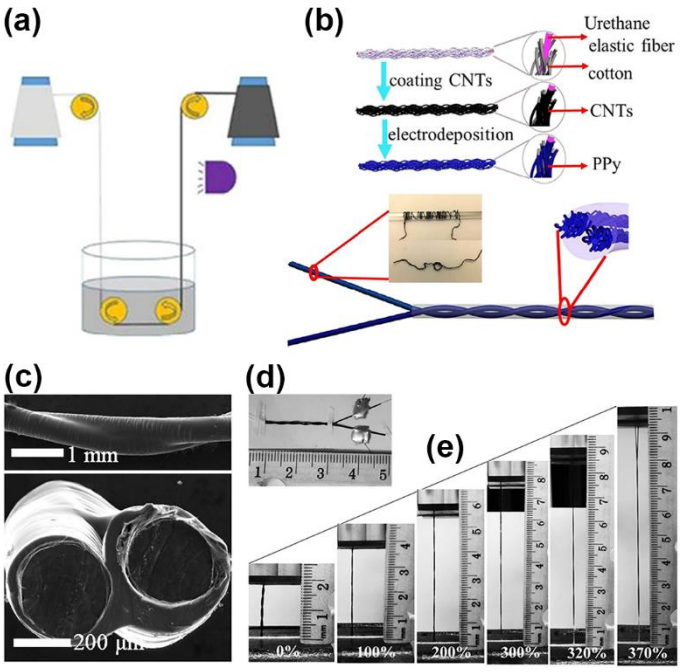

Figure 2. (a) Schematic showing CNTs coating and PPy electrodeposition on cotton/PU yarn and (b) assembly of yarn SC.[53] Copyright © 2016 Elsevier Ltd. (c) SEM images of the side view and cross-section of a twisted fiber SC. (d) Photographs of the stretchable fiber-based SC. (e) The stretchable fiber-based SC under different applied strain.[33] Copyright © 2015 WILEY-VCH.

The above examples demonstrate cost-effective and facile methods to produce stretchable fiber-based SCs with a combination of good stretchability and energy storage capability. However, they generally have low to moderate capacitance and energy density due to the insulating elastic core and thin layers of coatings. The mismatch in mechanical properties of the core and the coating also suggests the inevitable cracking or peeling-off of the coatings from the elastic core, which hinders the durability of the stretchable fiber-based SCs during long stretching cycles with large strains.

2.2 Structural engineering of non-elastic fibers

In the first approach discussed above, the stretchability of the fiber SCs is inherited from the elastic fiber used as the core. The second approach for fabricating fiber SCs is through structural engineering. It usually involves geometrical re-configuration of non-stretchable filaments into spring-like, helical or coiled structures, which are oftentimes achieved by twisting and/or coiling.

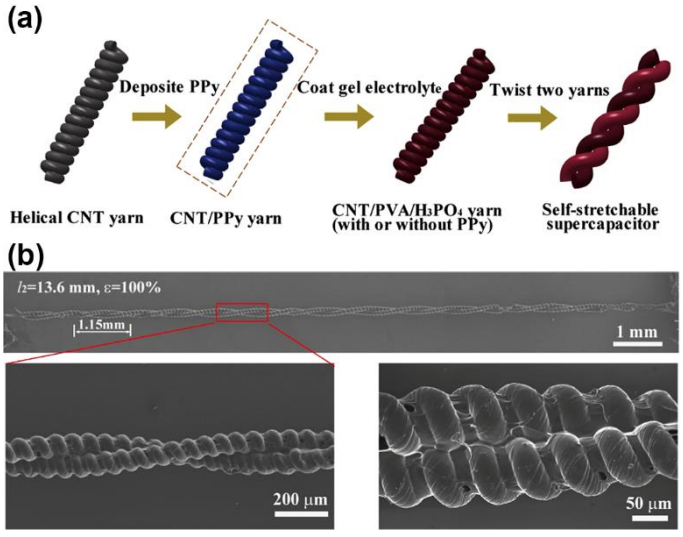

Figure 3. (a) Illustration of the fabrication process of stretchable fiber SC using structural engineering. A helical CNT yarn (with or without PPy on the surface) is coated by gel electrolyte, and two of such yarns are twisted into a double-helix supercapacitor. (b) SEM images of a twisted fiber SC with two

coiled fiber electrodes stretched by 100 %, showing uniform deformation of helical loops.[36] Copyright © 2014 Elsevier Ltd.

Twist insertion method was first reported by Shang *et al.* in 2012 as an effective method to prepare highly conductive and stretchable coiled CNT fibers.[54] Although these yarns could sustain large tensile strains of up to 285 % by opening the CNT loops, they only retained a stable electrical conductivity and stable spring constants under lower strain (*e.g.* 20–40%). Nevertheless, this work extended the potential application of CNT yarns in stretchable fiber electrodes for energy storage devices. Inspired by the idea, stretchable fiber SCs based on two spring-like CNT fibers with parallel or helical structures was reported (**Figure 3**).[36] In a report from Peng's group, a stretchable fiber SC based on two coiled CNT yarns showed a specific volumetric capacitance of 18.12 F cm^{-3}, an energy density of up to 0.63 mW h cm^{-3} and a power density of up to 37.74 mW cm^{-3}.[38] In addition, it retained 90% of its capacitance when stretched to 100% strain, and showed 94% capacitance retention after 300 stretching cycles with an applied strain of 100%, indicating that the fiber SC had a stable electrochemical performance and good durability during repeated stretching.

However, the relatively low specific capacitance and energy density of pure coiled CNT fiber still cannot meet the demand of practical flexible and wearable devices. Incorporation of other pseudocapacitive materials such as conductive polymers[36] and metal oxides[35] is a feasible approach to increase the capacitance of the stretchable CNT yarn. For instance, by decorating the spring-like CNT fibers with a thin PPy layer, capacitance of the stretchable fiber SCs increased from 11.8 F g^{-1} (pure CNT yarns) to 63.6 F g^{-1} after PPy deposition and was stable under extreme deformation conditions such as stretching up to 150 % as well as arbitrary shape deformations and high frequency dynamic stretching.[36] In another report, a stretchable asymmetric fiber-shaped SC was prepared by using PPy/CNT as negative electrode and MnO_2/CNT as positive electrode.[55] A specific capacitance of 60.44 mF cm^{-2}

at the scan rate of 10 mV s^{-1} was achieved which was well maintained during repeated stretching at 20% strain. Due to the extended potential window to 1.5 V, a high energy density of 18.88 μW h cm^{-2} was achieved. Incorporating stretchable CNT yarn with higher capacitance metal oxide was also explored. Choi *et al.* presented a core-shell stretchable fiber SC based on overtwisted CNT fibers with pseudocapacitive MnO_2 deposition.[35] The parallel fiber SC reached a capacitance of 34.6 F cm^{-3}, with 84% capacitance retention when it was stretched repetitively to a strain of 37.5%. These results suggest that using stretchable and conductive CNT yarns with high capacitive active materials is a promising alternative to the use of insulating elastic fiber.

However, the widely adopted core-shell structure has some limitations as well. One obvious disadvantage is that only limited surface of CNT fiber is utilized as an effective loading site for active materials, while the core of the fiber contributes little to the electrochemical processes. Later on, a biscrolling method was explored as a promising solution since this method can incorporate large amounts of guest materials (>90 wt%) into CNT yarn.[56] This method opened up the opportunities in further increasing the capacitance of stretchable CNT yarns by increasing the loading amount of capacitive active materials. Choi *et al.* produced stretchable CNT yarn supercapacitor with a high MnO_2 loading (70 wt%). This stretchable MnO_2/CNT yarn could be stretched to 320% and showed a capacitance of 382 mF cm^{-2} (17.7mF cm^{-1} and 105 F cm^{-3}). The stretchable SC fabricated with this yarn showed an energy density of 12 μWh cm^{-2} and a power density of 350 μWcm^{-2} which are much higher than other stretchable fiber-based SCs. This suggests that stretchable SCs based on biscrolled CNT yarns are especially promising considered their high capacitance and high energy density which originate from the high active material loading.

2.3 Structural engineering with stretchable substrate

Apart from using either stretchable substrate or structural engineering to create stretchability, there are also few reports that employed a combination of both approaches for the fabrication of stretchable fiber-based SCs. Xu *et al.* reported a stretchable fiber SC using twisted CNT fibers attached on a spandex fiber (**Figure 4**).[24] Two CNT yarns spun from a vertically aligned CNT arrays were twisted together and attached on a pre-strained spandex fiber. This fiber SC showed a capacitance of 4.99 mF cm^{-2}, a maximum power density of 0.49 mW cm^{-2} and an energy density of 0.23 μWh cm^{-2}. It could retain the initial capacitance after 10,000 charge-discharge cycles and 20 stretching cycles with an applied strain of 100%. Later, researchers from the same group developed asymmetric stretchable fiber SC based on a similar approach. By further precipitating pseudocapacitive MnO_2 particles on CNT yarn, the fiber SC showed an improved specific capacitance of 16.87 mF cm^{-2} (volumetric capacitance of 21.21 F cm^{-3}) and higher power density and energy density of 1.50 mW cm^{-2} and 1.14 μWh cm^{-2} respectively. This device also demonstrated a stable capacitance during 20 stretching cycles (at 100% strain) and 10,000 charge-discharge cycles.

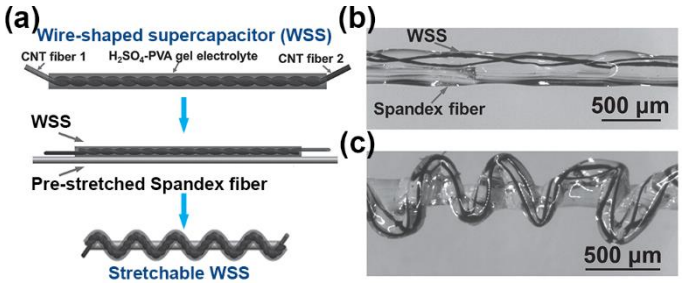

Figure 4. (a) Schematic of fabrication of a stretchable wire-shaped fiber SC by using an elastic fiber substrate, and optical microscopy images of an assembled fiber SC with Spandex fiber substrate at (b) pre-stretched and (c) relaxed state.[24] Copyright © 2013 WILEY-VCH.

Lu *et al.* developed PANi coated wet-spun CNT/graphene fiber as the electrode for fiber-based SC (**Figure 5**).[57] Two symmetric composite fibers were coiled on a stainless steel rod, and subsequently cast and embedded within a rubber matrix. Although the resulting fiber-

based SC was thick (millimeter scale), it showed an outstanding capacitance of 273.7 mF cm^{-2} (137.5 F g^{-1}) and 99.2% of its capacitance remained when stretched to 800% strain.

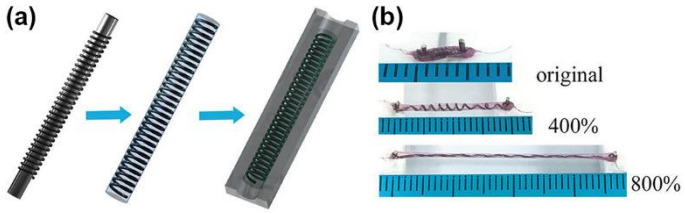

Figure 5. (a) Schematic illustration of the fabrication of a stretchable fiber SC. A yarn-like SC is coiled on a rod, the rod is then removed and the device is placed in a mold to be thoroughly covered with rubber. b) Illustration of prepared SC with different strains up to 800%.[57] Copyright © 2017 WILEY-VCH.

Another novel design has been developed for increasing the operating voltage of stretchable fiber-based SC. Wang *et al.* reported an "internal tandem" stretchable fiber SC, produced by wrapping PEDOT:PSS fiber on an elastic polymer fiber.[58] The fiber SC consisting of eight cells connected in series showed a high-voltage output of 12.8 V and ultrahigh energy density of 41.1 μW h cm^{-2} and power density of 3520 μW cm^{-2}. When the elastic SCs were tested under repeated stretching, 94% of their original specific capacitance remained after 200 stretching cycles with an applied strain of 300%.

3. Challenges and outlook

3.1 Challenges in design and fabrication

Despite significant progress in the area of fiber-based SCs, there is a lack of facile, efficient, and scalable fabrication of stretchable fiber-based SCs for textile-based wearable energy storage applications. There are still many challenges in the design and fabrication of stretchable fiber-based SCs, which will need to be addressed before they can be used for practical applications. Those limitations and challenges stem from the inherent rigidity and brittleness of the active materials used in making fiber electrodes, which do not meet the high

stretching requirements of stretchable fiber-based SCs. In order to achieve high-performance stretchable fiber-based SCs, it is critical to develop electrodes with high electrical conductivity and energy storage properties that once integrated into a fiber-based SC will lead to a highly stretchable device.

The mechanical mismatch of active material with elastic substrate made the traditional coating approach unsuitable to fabricate stretchable fiber-based SCs. To improve the adhesion of active materials to the stretchable elastic fiber substrate, some researchers have pre-stretched the fiber substrate and then coated or wrapped the active material. While researchers generally reported good adhesion using this approach, it is necessary to confirm that the active material layer would not peel off or develop microstructural cracks under repeated deformations as these would lead to device performance decay or failure. Moreover, the coating or wrapping approaches use stretchable substrates that do not contribute to the energy storage performance of the device. Researchers have oftentimes reported the energy storage performance of their stretchable devices in terms of mass, volume, or area of only the active material and did not take into account the substrate in their calculations. Once the mass, volume, or area of the whole device is considered, low energy storage performance is expected as coating or wrapping approaches typically achieve a thin layer of active material on the substrate. Alternatively, researchers have made spring-like stretchable fiber electrodes by winding fiber that contains active material on a flexible substrate. Although this approach can potentially incorporate higher amount of active materials than coating or wrapping, the devices made of spring-like SCs were relatively bulky and had large cross-section diameters that in some cases exceeded 1 mm.

Integrating active materials within stretchable polymer fibers also has its own issues; it generally leads to low conductivity and because of the presence of polymer layer around the active materials, which limits the access of electrolytes, low energy storage property is

expected. Composite approaches typically use low active materials loadings. Thus, the loading of active material will need to be maximized to achieve fiber electrodes with high energy storage properties. While stretchable conducting composite fibers have already been produced and successfully used for stretchable wires[42, 59] and wearable strain sensors,[43-46] their energy storage properties are still unexplored. Notably, the influence of active materials (loading and composition) on the energy storage properties of the stretchable composite fibers have not yet been investigated.

Structural engineering such as coiling has also been employed to impart stretchability to otherwise not highly stretchable fibers. While coiling typically achieves lower stretchability than coating or wrapping approaches, it has the advantage of eliminating the use of stretchable substrates and can utilize high active materials content. However, it is argued that the access of electrolytes to the materials located inside of the coils would be limited in this approach, resulting in ineffective usage of active materials.

The combination of structural engineering and elastic substrate has shown to greatly improve the stretchability of the fiber-based SCs. Elastic polymers were often used as either elastic substrate or a protective layer for the fiber-based electrode in these designs. Reports published recently suggested that the advantages of structural engineering and elastic substrate can be combined by rational design and careful engineering. However, this approach is relatively new and unexplored. Efficient and reliable design to fully utilize the performance of elastic polymer and fiber electrode are still lacking to date, which calls for further investigation and research.

3.2 Criteria and standard for performance evaluation

While some good progress has been achieved in fabricating suitable electrodes for stretchable fiber-based SCs, their electrical and energy storage properties are still far from the

requirements of wearable applications. High conductivity is particularly important in this case, because stretchable fiber-based SCs are expected to operate at a longer length than their non-stretchable counterparts. Fiber electrodes with low conductivities are known to lead to short devices because of increasing resistance with length, which results in low energy storage performance. Also, the energy storage performance of the stretchable fiber-based SCs should not change at various stretching levels. However, it is challenging to achieve constant electrical properties at different electrode lengths, and these conductivity changes can cause fluctuations of the device energy storage.

The lack of appropriate and standard testing procedures in the available literature has also made it difficult to compare the overall performance of the stretchable fiber-based SCs. Researchers have tested their stretchable fiber electrodes and SC devices at various conditions and did not report all relevant results. For instance, in some cases the performance of the stretchable fiber-based SCs has been shown at a much lower stretching conditions than the maximum reported stretchability. As a guide, we suggest that electrical and electrochemical properties of both fiber electrode and the SC device to be reported at various stretching levels. The electrode and the device should also be tested for their elastic recovery (*i.e.* the ability to recover to original length) and energy storage stability under cyclic deformations. Researchers should make reasonable efforts to make it clear whether they are reporting the energy storage performance in terms of mass, volume, or area of active material or the whole device.

From the manufacturing point of view, the fabrication of stretchable fiber-based SCs typically involves many tedious steps and have only been achieved in short lengths. Developing new fiber fabrication methods or creating novel fiber designs that can lead to improved electrical conductivity and energy storage of the fibers while retaining the fiber stretchability, opens up the possibility of scaled-up manufacturing. Researchers have assessed

the potential textile integration of their stretchable fiber-based SCs by manually weaving or stitching those into small pieces of textiles. While hand-weaving is useful for demonstration purposes, researchers should show their stretchable fiber-based SCs are compatible with conventional textile processing techniques, *e.g.* knitting or weaving. Here, the fiber electrodes or devices need to be tough enough to withstand the tensile and shear stresses and strains which are applied in textile processing. Moreover, individual stretchable fiber-based SCs possess limited output voltage and current which may not be sufficient for high energy needs of wearable devices. Thus, it is important to connect stretchable fiber-based SCs in series or parallel to enhance their performance. While possible to connect the devices at the textile processing stage, establishing the connections may be the most challenging task here.

Wearable applications also demand washability, safety, comfort, and aesthetic appeal. However, these important properties have been overlooked in literature not only for stretchable but for all fiber-based SCs. While it is possible to improve washability by encapsulating the stretchable fiber-based SCs with polymer coatings, it is not expected to be able to wash those like standard everyday clothes. Even different types of fabrics require their specific washing procedure and some can only be dry-cleaned such as wool suits and silk dresses. The stretchable fiber-based SCs can be made into patches and attached to the garment and detached at the time of washing. Flaking off potentially hazardous active materials (*e.g.* CNTs) and leaking out the electrolytes are two main safety concerns which become critically important once stretchable fiber-based SCs are to be used in contact with the wearer's skin. Appropriate packaging of the device might reduce the safety risks associated with using stretchable fiber-based SCs. Further research is required to improve comfort of the stretchable fiber-based SCs by means such as decreasing the fiber diameter, improving their flexibility, and texturizing processes. Current stretchable fiber-based SCs can only be made in dark colors. Their aesthetic appeal can be improved by investigating the

possibility of dying into different colors or by incorporating them with other yarns. While no reports have carried out cost evaluations, due to the use of expensive active materials and processes, it is expected that the textiles made of stretchable fiber-based SCs to be costly. Therefore, low-cost materials and fabrication approaches should be used in order to make stretchable fiber-based SCs economically viable.

3.3 Summary and outlook

Stretchable fiber-based SC offers viable energy storage solutions for a wide range of wearable, portable, and integrated devices. The guidelines and directions provided in this feature article can help researchers to come up with solutions to the existing challenges of this exciting field and achieve highly stretchable fiber-based SCs with stable and high energy storage properties, which can be produced in large-scale and integrated into fabrics using textile processing for practical use. The successful development of stretchable fiber-based SCs requires strong collaboration between specialists in material science, electronics, textile and fashion. Advancements in materials frontier can open up the possibility of synthesizing and processing active materials that meet the stringent requirements of stretchable fiber-based SCs, while electrical engineers can help maximize the energy storage performance and devise the circuits that connects stretchable fiber-based SCs to electronic devices. Textile and fashion designers can make sure that the stretchable fiber-based SCs are integrated within garments in such way that achieves wearability, comfort, and aesthetic appeal while maintaining the energy storage performance.

Table 1. Summary of the stretchable fiber-based SC reported in literature.

Elastic structure fabrication techniques	Electrode fabrication techniques	SC structure	Materials	Active materials	Conductivity/Resistivity of electrode	Specific capacitance of device at 0% strain	Energy density/Power density of device at 0% strain	Max strain	Capacitance retention (at specific strain)	Ref.
Wrapping on elastic substrate	Wrapping aligned CNT sheets on elastic fibers	**Coaxial**	CNT/rubber	CNT	--	[a]19.2 F g^{-1}	0.515–0.363 Wh kg^{-1} 19–421 W kg^{-1}	100 %	95% (after 100 stretching cycles, 75 % strain)	[34]
	Wrapping aligned CNT sheets on elastic fibers and coating with PEDOT:PSS	**Twisted**	CNT/PEDOT:PSS/rubber	CNT/PEDOT:PSS	--	[a]30.7 F g^{-1}	--	370 %	~100% (after 100 strain cycles, 200% strain)	[33]
	Wrapping CNT sheet deposited with PANi on elastic polymer fiber	**Coaxial**	CNT/PANi/elastic polymer	PANi	--	[a]111.6 F g^{-1} [a]50.1 F cm^{-2}	--	400%	~71% (after 5000 strain cycles, 300% strain)	[32]
	Wrapping CNT/PANi sheets on rubber fiber	**Parallel**	CNT/PANi/ rubber fiber	PANi	--	[a]255.5 F g^{-1}	--	100 %	97 % (after 100 strain cycles, 100% strain)	[61]
	Wrapping PEDOT:PSS fiber on elastic fibers	**Coaxial**	PEDOT:PSS/	PEDOT:PSS	1771.8 S cm^{-1}	93.1 mF cm^{-2} 74.5 F cm^{-3}	41.1 μWh cm^{-2} 3520 μW cm^{-2}	400 %	94 % (after 200 strain cycles, 300% strain)	[58]
	CNT coated opposite side of silicon ribbon and the n deposited with MnO_2 and PEDOT on CNT	**Paralle**	MnO_2/PEDOT/CNT/silicon ribbon	MnO_2/PEDOT/	179 Ω cm^{-1}	2.38 mF·cm^{-1} 11.88 mF·cm^{-2} 158.4 mF·cm^{-3}	5.5 uWh·cm^{-3}	40 %	92.8% (after 1000 strain cycles, 200 strain)	[30]
Structural engineering	Biscrolled MnO_2/CNT into helical fiber	**Parallel**	MnO_2/CNT	MnO_2	170– 29 S cm^{-1}(at 0% strain)	17.7 mF cm^{-1} 382 mF cm^{-2} 105 F cm^{-3}	--	~320%	41.6% (after 100 strain cycles, 30 strain)	[60]
	Wrapping the CNT/MnO_2 sheet ribbons on the nylon fibers and then twisted to coil fiber electrode	**Parallel**	Coiled MnO_2/CNT/nylon fiber	MnO_2	0.4 kΩ cm^{-1} (at 0% strain) 2.8 kΩ cm^{-1} (at 150% strain)	[a]5.4 mF cm^{-1} [a]40.9 mF cm^{-2}	[a]2.6 μWh cm^{-2} [a]66.9 μW cm^{-2}	150 %	81.6% (150 % strain)	[37]
	twisting CNT fiber into helical structure	**Parallel**	CNT	CNT	0.19 kΩ cm^{-1} (at 0% strain) 0.25 kΩ cm^{-1} (at 100% strain)	18.12 F cm^{-3}	0.629 mWh cm^{-3} 37.74 mW cm^{-3}	100 %	94% (after 300 strain cycles, 100% strain)	[38]
	Deposited MnO_2 on coiled CNT fiber	**Parallel**	Coiled MnO_2/CNT Core–Shell Structure Yarn	MnO_2 /CNT	--	26.5 F g^{-1} 2.72 mF cm^{-1} 61.25 mF cm^{-2} 34.6 F cm^{-3}	2.4 mWh cm^{-3}	37.5%	95% (after 1000 strain cycles, 20% strain)	[35]
	electrochemical deposition of MnO_2 and PPy on CNT fiber	**Coaxial**	CNT/MnO_2 fiber/CNT/PPy	MnO_2 and PPy	--	60.43 mF cm^{-2} 7.72 F g^{-1} 9.46 F cm^{-3}	18.88 μW h cm^{-2} 2.41 mW h g^{-1} 2.98 mW h cm^{-3}	--	88% (after 200 strain cycles, 20% strain)	[55]

						19.86 mF cm^{-1}	6.206 μW h cm^{-1}			
	Electrochemical coiled CNT fiber deposition of PPy on	**Twisted**	CNT/PPy	PPy	--	38.1 F g^{-1}	--	50 %	96% (after 10000 strain cycles, 50% strain)	[36]
	Electrodeposit graphene on graphene fiber and wrapped on template to form spring structure	**Twisted**	graphene core–sheath fibers	Graphene	10–20 S cm^{-1}	18 μF/cm 1.2 mF/cm^2	--	200 %	--	[17]
Combination of methods	Chemical deposited MnO_2 on the CNT fiber and gel electrolyte	**Twisted**	MnO_2/CNT hybrid fiber/CNT fiber	MnO_2/CNT	--	157.53 μF cm^{-1}	46.59 nWh cm^{-1} 56.16 μW cm^{-1}	100 %	--	[23]
	Glued CNT fibers on elastic fibers	**Twisted**	CNT/spandex fiber	CNT	--	4.28 mF cm^{-2}	1.32 W cm^{-3} 0.6 mWh cm^{-3}	100%	97% (after 20 strain cycles, 100% strain)	[24]
	Electrodeposit PANi onto CNT/graphene fiber and wrap the supercapacitor as a spring structure, then mounted in elastic polymer	**Twisted**	Spring-structure CNT/graphene /PANi fiber	CNT/graphene/PANi	--	137.5 F g^{-1} 10.3 mF cm^{-1} 273.7 mF cm^{-2} 91.2 F cm^{-3}	--	Over 800%	~77% (after 5000 strain cycles, 800% strain)	[57]
	CNT film coated on twisted CNT fiber	**Coaxial**	CNT	CNT	10^2–10^3 S cm^{-1} (at 0% strain)	[a]59 F g^{-1} [a]32.09 F cm^{-3} [a]29 μF cm^{-1} [a]8.66 mF cm^{-2}	1.88 Wh kg^{-1} 755.9 W kg^{-1}	100 %	81.6% (after 75 strain cycles, 10% strain)	[62]
	Electrochemical deposition of PPy on co-spun cotton fiber and urethane elastic fiber coated with CNT	**Paralle**	cotton fiber/ urethane elastic fiber /CNT/PPy	PPy	--	69 mF cm^{-2}	6.13 μWh cm^{-2} 0.4678 mWh cm^{-3} 0.133 mW cm^{-2} and 10.18 mW cm^{-3}	80 %	85% (after 1000 strain cycles, 40% strain)	[53]
	MnO_2/oxidized CNT fibers mounted on PDMS film	**Paralle**	MnO_2/oxidized CNT	MnO_2	331–67 S cm^{-1}	409.4 F cm^{-3} 133 mF cm^{-2}	14.2 mWh cm^{-3} 6250 mW cm^{-3}	40 %	~100% (after 50 strain cycles, 40% strain)	[31]

[a] Calculated based on active materials without the substrate

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