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REVIEW

Buckled Fiber Conductors with Resistance Stability under Strain

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Abstract
The availability of fiber conductors that can be stretched to large extents without significantly changing resistance or conductivity could enable the advances of elastic conductors as electronic interconnects, electronic skins, stretchable sensors, wearable systems, and medical robots. Therefore, the preparation of fiber conductors with high stretchability is crucial to the development of flexible electronic devices. This review summarizes the advances in constructing fiber conductors with an emphasis on recent developments of buckled structural design, fabrication methodologies, and strategies, with the ultimate goal of achieving good stability of resistance or conductivity at large strains. This review classifies the buckled fiber conductors into inner buckling and outer buckling, and related examples are summarized, providing a context that buckled fiber conductors are geared towards applications in electrical interconnects, wearable systems, and smart medical robotics. The present challenges in this area are critically evaluated and our perspectives for improving the performance of the buckled fiber conductors for future applications are presented.

Keywords Buckled · Fiber conductor · Stretchability · Resistance

Introduction

With the miniaturization and high integration of electronic devices, electronic products have become more portable to promote the development of wearable devices [1–11]. Wearable electronic devices are portable with the movement of the human body, including biomedical sensors [12–18], wearable heaters [19–24], human–machine interactors [25–29], intelligent prostheses [30–34], and so on. It can be worn directly on the body or integrated into clothes or accessories. These wearable devices can adapt to different body types through deformation, and work normally when the body moves freely, such as walking, running, jumping, etc. [35–39]. With the advances of textile technology, stretchable fiber conductors have become crucial components of wearable devices [40–45]. The fiber diameter ranges from tens to hundreds of microns, and it has the advantages of being lightweight and stretchable. Fiber conductors already have applications such as energy harvesting [46–52], energy storage [53–57], sensors [58–63], and electrical actuators [64–68]. These electronic devices require stretchable fiber conductors as electrical interconnects. Traditional cables are not stretchable and cannot meet the needs of wearable devices. Moreover, there is an urgent need for a stretchable fiber conductor that maintains a constant resistance or conductivity upon stretching [69, 70]. Conductive films or fibers without buckled structure normally cannot resist larger deformation while maintain a stable electrical performance. Currently conductive materials such as metal fibers, CNTs, conductive polymers, graphene and other materials have limited stretchability. The purpose of designing the buckling structure inside the elastic conductive fiber or on the surface is to increase the stretchability of the fiber conductor while maintain the electrical performance.

It is known that the diameter of the fiber will reduce when it is stretched, which will lead to an increase in fiber resistance. Thus, to obtain resistance-stable fiber conductor under large strains has long been a challenging problem. Previously, a stretchable fiber conductor was constructed by encapsulating liquid metal [71] in elastic rubber tubes. Though they can achieve stable conductivity under large strain, the potential safety hazards of these fiber conductors (material leakage at large strain) and the durability of the material (resistance change at large strain) limit its development towards application such demonstrated as charge

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cables. On the other hand, generating composite fibers with a buckled conductive film on the surface or inside of the fiber has become the mainstream solution for current stretchable fiber conductors. These fiber conductors are light-weight, simple in manufacturing, and have good durability and repeatability. The buckling phenomenon is used to guide the construction of these stretchable fiber conductors in different ways. However, the fiber-based buckling mechanism and its structural design rules, as well as manufacturing methods are not yet summarized. Therefore, this review will start with the formation of buckled structures, and will mainly discuss the advantages, structural features, and manufacturing methods of buckled fiber conductors. Future applications, challenges and perspectives for buckled fiber conductors are also presented.

Formation of Buckled Structures

The buckling mechanism of a film on an on a substrate is well discussed in the literature[72–77]. Here, we first introduce the buckling behavior of a two-dimensional bilayer structure, then describe the advances in how to construct fiber conductors with buckled structures[72]. As show in Fig. 1a, compressing a thin film on a pre-strained substrate can lead to buckling instability[73]. The thin film has a large Young's modulus, $\boldsymbol{E_f}$, which is difficult to deform in-plane. However, the film thickness , $\boldsymbol{h}$, is much smaller than the substrate. Its bending stiffness, $\boldsymbol{D} = \boldsymbol{E_f h}^3/12$, is very small compared to its modulus. Therefore, the film usually undergoes out-of-plane wrinkles and deformation, and the internal pressure stress is released by generating bending energy. We use simple illustrations (Fig. 1a) to describe the four stages of a buckled film on a soft substrate. Figure 1a(i) shows that the top conductive layer adheres to the substrate, under none to minor compression, the top layer does not buckle and remains flat. With the increases of compression, the top layer buckles into small waves on top of the elastic substrate but does not delaminate from the interface. The buckled surface reduces the system energy of the film. The interface between the top layer and the elastic substrate usually has defects, and the degradation of the interface will reduce the adhesion. Under further compression, many small buckled structures converge to form a large buckled structure and the phenomenon of local separation from the substrate occurs, which is the partial delamination mode. This phenomenon will continue as the compression force increases. Until the film is completely separated from the surface of the soft substrate [74].

The buckling formation mechanism of thin films can be correlated to formation of fiber conductors with buckled structures. The conductive strips or films can be respectively designed to inside of a hollow elastic fiber or outside of a solid elastic fiber, generally presenting a coaxial structure of fibers. If the stretchability of the elastic fiber substrate

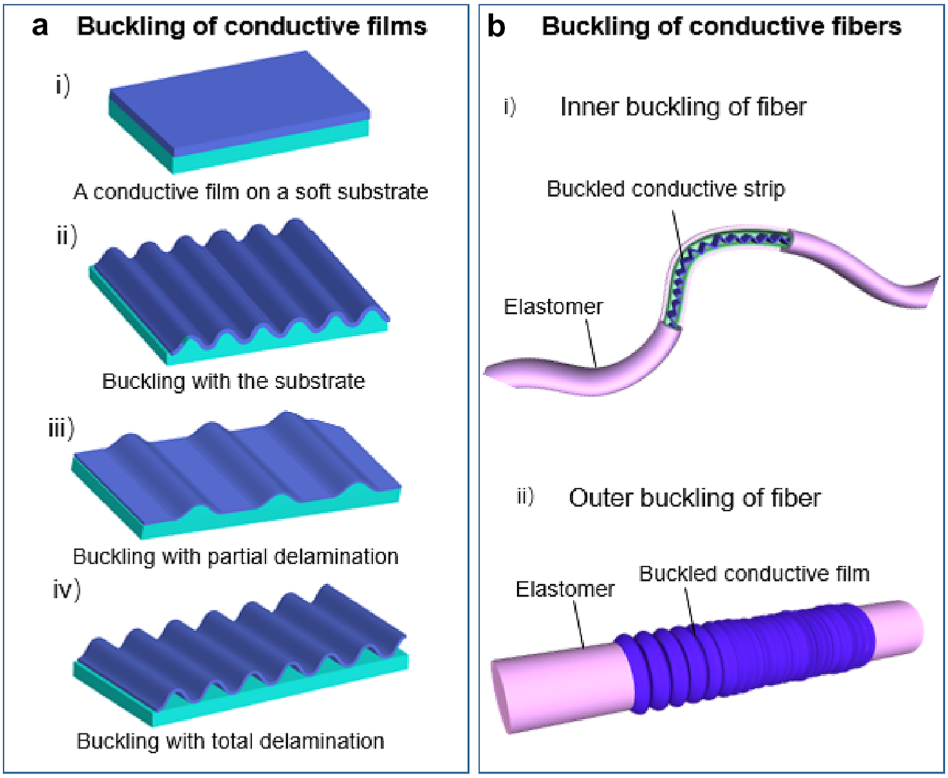

Fig. 1 **a** Schematic illustration of the buckling structure for a thin film on a flexible substrate. (i) Wrinkle-free bilayer structure. (ii) Surface buckling with no delamination. (iii) Buckling with a partial delamination. (iv) Buckling with a total delamination. **b** Buckled structure of the conductive fiber. (i) Inner buckling of fiber (top) and (ii) outer buckling of fiber (bottom)

exceeds 1000%, larger compressive stress is generated when the elastic fiber relax from the stretch state, which is beneficial to build the high-density buckling structure. According to the structural characteristics of fiber conductors, it is divided into an inner buckling type and outer buckling type, as shown in Fig. 1b. These fiber conductors require multimaterials to play different roles, and could be stretched until the buckled films become straight. In the case of inner buckling mode, the buckled and conductive strip is usually delamilated from the outer elastic tube, presenting a unique free standing, buckled structure. Thus, the effective total length of the conductive parts remains constant unpon stretching, which is the key concept for coaxial, and elastic fiber conductors that can resist resistance change under larger strains. Thus, before the failure of mechanical extension, the resistance of the fiber will be stable. In an outer buckling mode, highly stretchable coaxial fiber conductors can be fabricated by wrapping conductive nanosheets on stretched rubber fiber cores. It was efficient in creating fiber with a stretch-insensitive resistance at very high strains by introducing hierachichally buckled structures, but with a sacrifice to expose conductive carbon nanomaterials.

Structural Design and Fabrication of the Fibers

Structural design is very important to achieve good stretchability of the fiber conductors. We introduce the structure and performance of fiber conductors with inter buckling and outer buckling examples and briefly summarize the materials and structures, performance, and key fabrication methods in Table 1.

Figure 2a–d show examples of the inner buckling of fibers. Zhou et al. reported a buckled conductive polymer ribbons in elastomer channels as a stretchable fiber conductor through a combination of coaxial wet-spinning and solution stretching-drying-releasing process. The core layer of this fiber conductor is a conductive composite of a conductive polymer, (poly (3,4-ethylene dioxythiophene)/polystyrene sulfonate (PEDOT/PSS)) and a copolymer (polyethylene-block poly(ethylene glycol) (PBP)). The outer layer material is a thermoplastic elastomer. Skyscan CT (Fig. 2a) images and SEM images (Fig. 2b) show the morphology of the inner buckling conductive film varies with different fabrication pre-strains. Under low restrain, the conductive film buckles randomly inside, while under high pre-strains, the internal conductive film is closely buckled and stacked. Figure 2c shows the relative change in resistance,$\Delta R/R_0$, of the coaxial fiber under different strains, presenting the relative resistance changes of the fiber are less than 4% under 680% strain [78].

Figure 2e–h shows examples of the outer buckling of fibers. The fiber conductors with a sheath-core structure were prepared by wrapping highly oriented carbon nanotubes (CNTs) aerogel on a stretched rubber fiber (Fig. 2e). The CNTs are arranged perpendicular to the surface of the rubber fiber and form a multi-level buckling structure after releasing the applied strains. This method gives the fiber a maximum stretchability of 1320%, and the relative resistance changes less than 5% under 1000% applied strain [79]. Zhang et al. [84] designed a core-sheath stretchable conductive fiber that can work in water. The fiber starts from pre-strained Lycra fiber followed by spray coating one-dimensional conductive CNTs/silver nanowires (AgNWs) and wrapping styrene-(ethylene-butylene)-styrene (SEBS) thin film. Figure 3f shows the longitudinal section structure of the fiber, composed of polyurethane (PU) core fiber (light green) and conductive sheath. The left is the SEM image of the fiber surface, and the right is the SEM image of AgNWs

Table 1 Summary of the properties and preparation methods of stretchable conductive fibers with buckled structures

Material and buckling structure	Conductivity and resistivity	Max mechanical steatchability& highest tensile strain of conductivity or resistance stable value	Key fabrication methods	Ref
Metal and AgNW; Elastic channel buckling	–	100%; 300%	dip-coating and pretraining-releasing	[66]
PEDOT/PSS; Elastic channel buckling	7.8 S cm^{-1}	680%; 900%	Wet-spinning and solution stretching-drying-releasing	[78]
CNT; sheath-core structure	3.6 S cm^{-1}	1000%; 1320%	Pretraining- CNT wrapping-releasing	[79]
MWCNT; sandwich structured	(180 Ω· cm^{-1})	200%: -	Pretraining- CNT wrapping-releasing	[80]
CNT; supercoil structure	–	1000%; 1500%	Pretraining- CNT wrapping-Twisting-releasing	[81]
CNT; supercoil structure	–	600%; 800%	Pretraining- CNT wrapping-Twisting-releasing	[82]
Graphene; caterpillar structure	1.24S cm^{-1}	815%; 1010%	Coating and pretraining-releasing	[83]

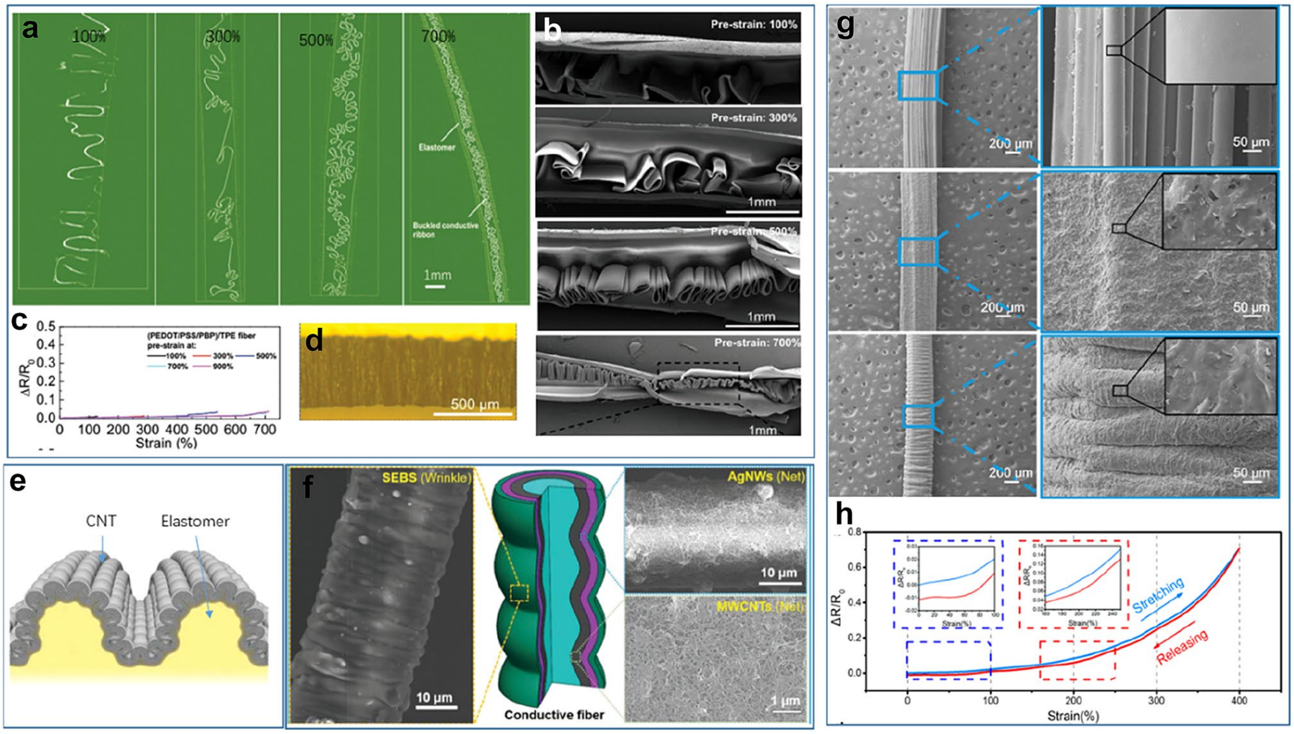

Fig. 2 Buckled structural designs of fibers. **a** Skyscan CT images of the buckled ribbons in the elastomer sheath with different fabrication pre-strains. **b** Cross-sectional SEM images of the coaxial fiber fabricated without pre-strain process and SEM images of buckled PEDOT/PSS/PBP ribbons inside the TPE channel at 100%, 300%, 500%, and 700% pre-strain. **c** Resistance change diagram under different stretching ranges. **d** Picture of the conductive film under an optical microscope. Reproduced with permission [78]. Copyright 2019, WILEY–VCH. **e** Illustration of the structure of a longitudinal section of the CNT. Reproduced with permission [79]. Copyright 2015, SCIENCE. **f** SEM image of fiber surface structure (left) and inner layers of AgNWs and MWCNTs (right)Longitudinal section structure of fiber (center). Reproduced with permission [84]. Copyright 2019, WILEY–VCH). **g** SEM image of graphene-free, 0-layer, and 300-layer PU fibers. **h** Resistance change graph under different stretch ratios Reproduced with permission [83]. Copyright 2019, Nano Letters

and MWCNTs of the conductive sheath. The fiber exhibits a stable core conductivity ($R_0 \approx 2 \times 10^4$ S m^{-1}, $\Delta R/R_0 \approx 0.1$ at 100% strain). The thickness of SEBS can be adjusted to protect the skin from the exposure of CNT and AgNWs. Sun et al. [83] used graphene material to coat the surface of the pre-stretched PU fiber (Fig. 2g) and designed a conductivity stable fiber with a worm structure. Figure 3g shows the SEM image of graphene-free, 0-layer, and 300-layer graphene-covered PU fibers. With 300 layers of graphene, the graphite sheath shows a clear buckling structure. Figure 3h shows the reversible relative resistance change of a typical 300-layer graphene fiber, showing excellent resistance stability at 400% strain. Moreover, the fiber exhibits strain-insensitive characteristics ($\Delta R/R < 0.1$) under a strain of less than 220%, which is significant for the communication stability of wearable devices.

Other than typical outer buckled structures of fibers, twistable, and stretchable fiber conductor with sandwich structures were also reported (Fig. 3a). The CNT electrodes are sandwiched on both sides of the insulating rubber core layer. In the process of fiber stretching, the CNT on both sides can absorb the sheer force brought by the deformation process to provide a constant electrical conductivity. Besides, the sandwich fibers provide the functions of strain sensing, by generating capacitance changes during stretching (200%) and giant twist (1700 rad·m^{-1} or 270 turns·m^{-1}), respectively. This feature can be used for strain sensing and fiber energy devices [80]. Microscopically buckled coiled Fibers composed of coiled and pre-stretched rubber core layer wrapped with CNT film. Figure 3b is a schematic diagram of the fiber structure (top) and the morphology under an optical microscope (bottom). The fiber shows a stable electrical conductivity at 400% strain with a 10-layer CNT [81]. Moreover, a fiber with a supercoiled structure can reach an ultra-high stretchability up to 1500%. The fibers present a highly ordered and dense structure along the fiber direction. When stretched to 1000% strain, the relative electrical resistance of the supercoil fiber increases by 4.2%, revealing excellent resistance stability at very large strain. Figure 3c shows the model of the supercoil fiber (left) and the SEM images of the fiber in three crimped states during the preparation process (right), indicating that the stretchability of

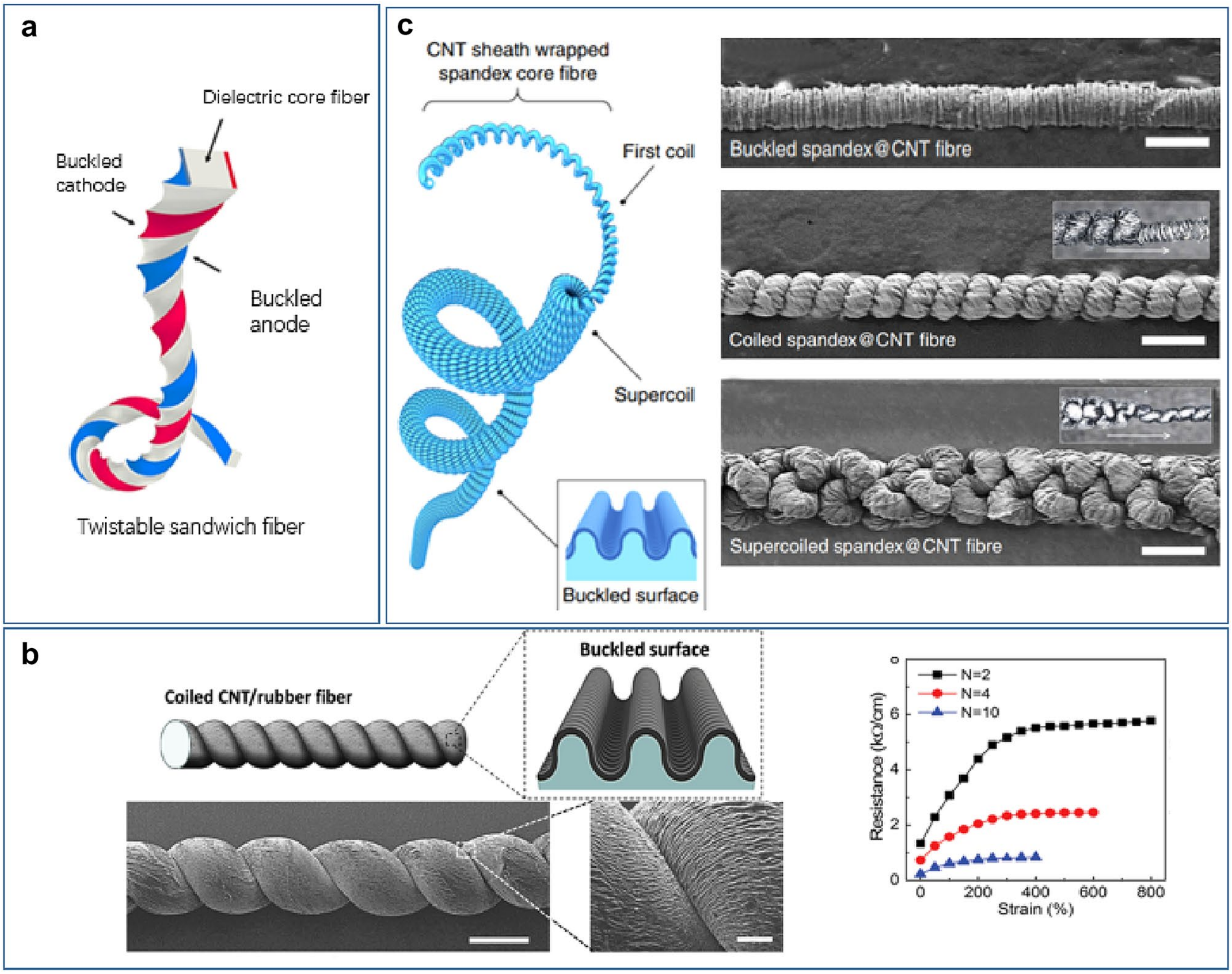

Fig. 3 Buckled structure of twisted fiber conductors. **a** Schematic illustration of a twist-inserted rectangular sandwich fiber. Reproduced with permission [80]. Copyright 2016, Nano Letters. **b** Schematics illustration of the highly twisted spandex@carbon nanotube fiber, consisting of first-coils and supercoils. Reproduced with permission [81]. Copyright 2019, Nature communication. **c** Schematic illustration of the surface of buckled electrode fiber. Reproduced with permission [82]. Copyright 2016, WILEY–VCH

the fiber can be further improved through over-twisting and knitting technologies [82].

At present, conductive fibers with buckling structures are constructed with a typical “prestrain-release” concept. Figure 4a shows the process of “solution stretching-drying-buckling” for preparing a buckled conductive film in an elastic channel. After the coaxial fiber is prepared by wet spinning, the solvent in the conductive polymer dispersion volatilizes from the porous TPE material to form a conductive film. After releasing the pre-stretched fiber, the inner dried films were compressed by the TPE sheath and buckled structures are obtained, leading to resistance and conductivity stability under large tensile strains. Figure 4b shows the preparation process of PU/graphene conductive fibers. The original PU filaments are pre-stretched and immersed in graphene/PU ink for absorption, and the PU fibers form a graphite microlayer on the PU filaments impregnated in the coagulating water bath. After releasing the pre-strain applied on the PU filaments, a worm-shaped graphene microlayer can be obtained. Figure 4c schematically illustrates the preparation process of a buckled and coiled fiber. First, the silicone rubber fiber prepared by twisting a capillary tube, and then a pre-strain is applied to the rubber fiber and then wrapped with CNT thin film. Finally, buckled coiled fiber is obtained after releasing the pre-strain. Figure 4d shows the fabrication of core-sheath stretchable conductive fiber, which is composed of PU fiber as the core and the sheath layer. The addition of metallic AgNWs is to further enhance the conductivity of the fiber. The fiber was prepared by pre-straining the PU fiber, then the MWCNT layer and the AgNWs layer are sprayed on the pre-stretched fiber surface, then a SEBS layer is coated. The SEBS layer

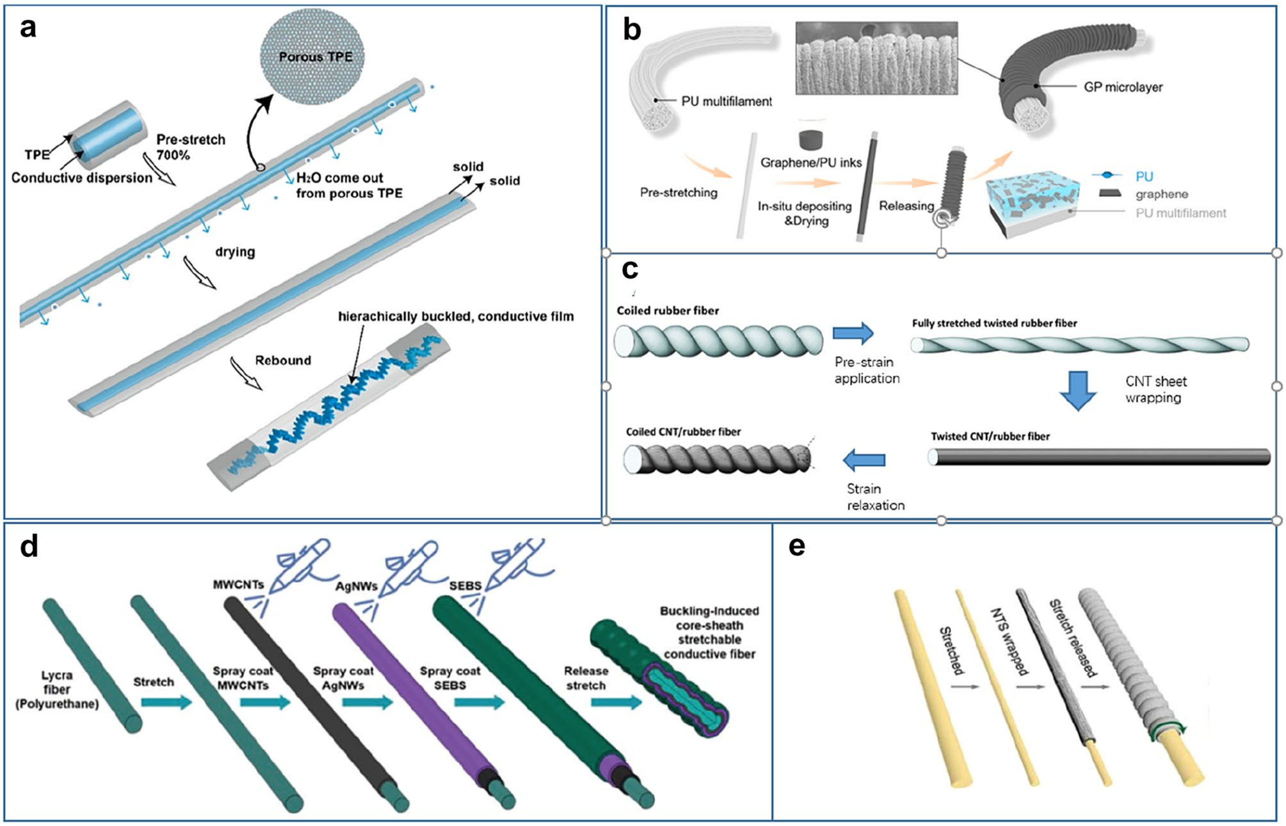

Fig. 4 Fabrication methods to create buckled fiber conductors. **a** Schematic illustration of PEDOT/PSS PBP fiber, using the "solution–stretching–drying–buckling" method. Reproduced with permission [78]. Copyright 2019, WILEY–VCH. **b** Schematic diagram of preparing the worm-shaped fibers [83]. Reproduced with permission. Copyright 2019, Nano Letters. **c** Schematic illustration of the fabrication of a buckled electrode fiber. Reproduced with permission [81]. Copyright 2016, WILEY–VCH. **d** Schematic diagram of the fabrication process of core-sheath stretchable conductive fiber. Reproduced with permission [84]. Copyright 2019, WILEY–VCH). **e** Schematic diagram of the preparation of sheath-core fibers. Reproduced with permission [79]. Copyright 2015, Science

is coated as the outer protective material, which can give the fiber waterproof, as well as wear-resistant properties. Figure 4e shows the fabrication process of the hierarchically buckled sheath-core conductive fibers. First, the rubber fiber core was stretched, then wrapping of CNT aerogel film as the sheath. The orientation of individual CNTs was perpendicular to the rubber fiber direction. Finally, the hierarchically buckled sheath-core fiber was obtained after releasing the pre-strain. In summary, the "prestrain-release" concept is generally used as the key step and mainstream to construct either inner or outer buckled layers of the fibers. The fabrication methods based on this concept is versatile to create buckled fibers that can meet different needs. Yet, in the future, a continuous process is needed to make continuous fiber conductors that possess resistance/conductivity stability at large strains.

Future Applications

Flexible and stretchable electronics are widely used in flexible displays [85–87], electronic skins [88–90], flexible sensors [91–94] and bio-electronic devices [8, 95]. Flexible electronic devices need the fiber conductors that maintain good conductivity under different strain to connect devices. Therefore, the preparation of elastic fiber conductors has become the key to the development of flexible electronic devices. The traditional method is to make the wire into a spring-like structure for connecting wearable devices [96]. However, the distribution of the thread increases, when bending joints such as the wrist, slack, or tangles tend to form. At the same time, it will cause inconvenience to the wearable device and affect the appearance of the device and the comfort of the wearer. One of a commercial stretchable cable was made by winding traditional copper cables on rubber (Fig. 5a left), and protected with textile covering, but its stretchability is less than 40%. Traditional stretchable cables could soon be replaced by inner buckled fiber

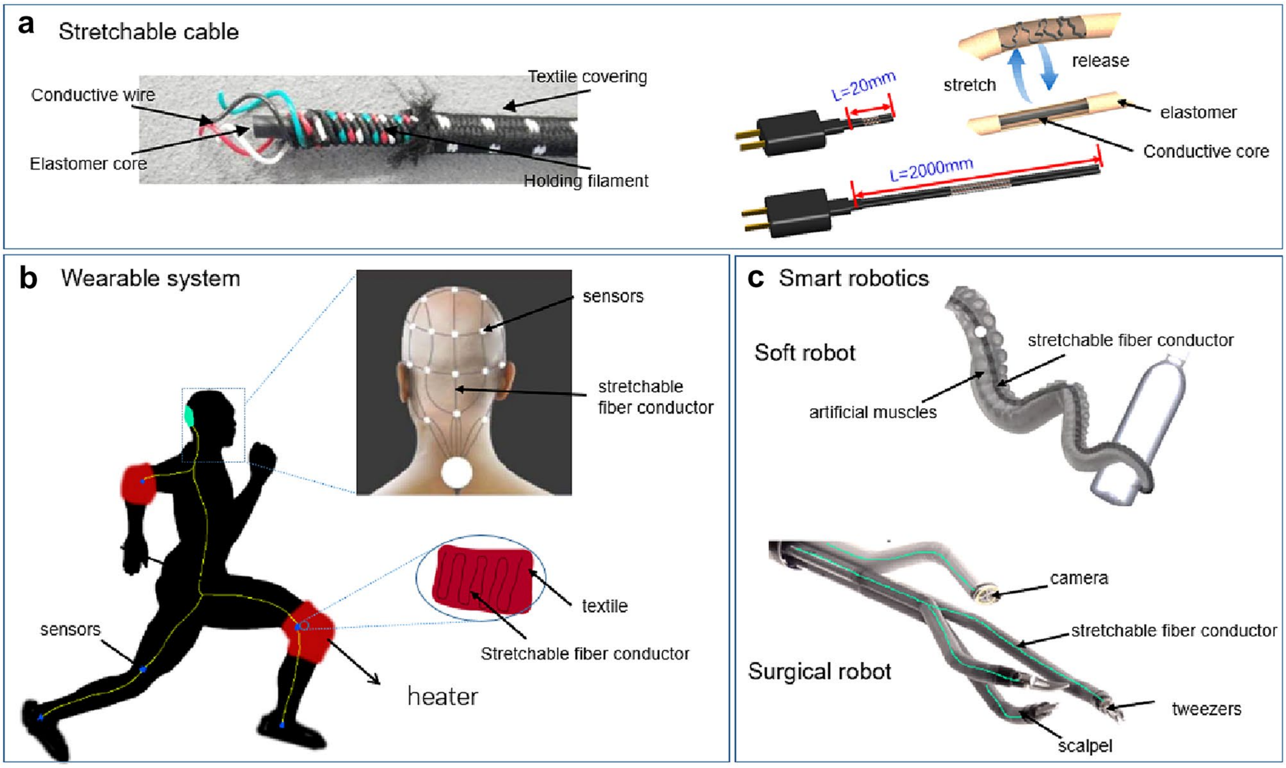

Fig. 5 Applications of stretchable fiber conductors. **a** Stretchable cables for mobile devices (charging cables, earphone cables). **b** application of stretchable fiber as a communication cable and heating component in wearable systems. **c** Application of stretchable fiber as a communication cable in the smart robots

conductors, features a stretchability of 680% with relative resistance change less than 4%. It can meet the stretching requirements of most weak current devices and possibly save space for the consumable electronics such as phone chargers (Fig. 5a right).

The flexible, wearable system is a new integrated system, which integrates human–computer interaction equipment, wearable heaters [97], artificial muscles, etc. Based on the characteristics that the stretchable fiber conductors can withstand large deformation and is suitable for complex surfaces such as head, joints, they can be widely used in wearable sensing systems as electrical interconnects or as heating elements in wearable heaters (Fig. 5b).

Stretchable fiber conductors are also the key to breakthroughs in robotics technology. Soft robots [98–100] (Fig. 5c) can bend, twist, and grab objects more than 100 times their weight, which stretchable fiber conductor makes a great contribution to the flexibility, conductivity, and toughness. They can also replace humans to finish dangerous tasks such as defusing a bomb and putting out the fire. Microrobots [101] also need stretchable fiber conductors. Microrobots are also a new current of medical equipment with a diameter of about 2 mm which are known as “The Never Tremulous Hands”. The use of microrobots can reduce the wound area of patients and can accurately complete the surgery even with slight disturbance thanks to the stretchable fiber conductor featuring fast conduction and precise operation.

Outlook and Conclusion

In summary, buckled fiber conductors that can resist large strains without a dramatic change in resistance or conductivity are crucial for next-generation flexible or stretchable electronics. The majority of current conductive materials for buckled fiber conductors are CNTs, graphene, metal nanowires, and conductive polymers. However, the intrinsic, non-stretchable nature of these materials has hindered the widespread use of fibers. Secondly, the conductivity of fibers is much lower than that of metallic wires or fibers, thus they cannot meet the normal working requirements of electronic devices. Most reported fiber conductors are prepared at a lab scale, and a continuous preparation route should be considered. To realize the commercial product of fiber conductors, the development of mass production technology to create buckled structure in-situ is essential. Besides, fiber conductors with exposed conductive materials are not compatible with mature textile technologies, which also requires researchers to develop new textile encapsulation technologies and unify smart textile standards. What is more, it is difficult to compare the reported performance of fiber conductors because of the lack of appropriate evaluation systems. In addition to conductivity, stretchability, and resistance stability at certain strain levels, other data such as cyclic mechanical or electrical repeatability should also be reported.

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Compliance with Ethical Standards

Conflict of interest The authors declare that they have no conflict of interest.

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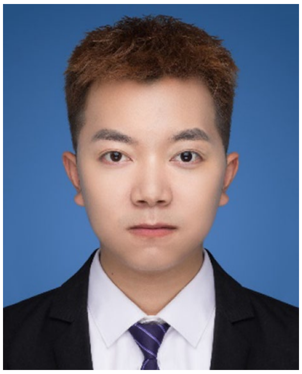

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