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Topical Review

Weavable thermoelectrics: advances, controversies, and future developments

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Abstract

Owing to the capability of the conversion between thermal energy and electrical energy and their advantages of light weight, compactness, noise-free operation, and precision reliability, wearable thermoelectrics show great potential for diverse applications. Among them, weavable thermoelectrics, a subclass with inherent flexibility, wearability, and operability, find utility in harnessing waste heat from irregular heat sources. Given the rapid advancements in this field, a timely review is essential to consolidate the progress and challenge. Here, we provide an overview of the state of weavable thermoelectric materials and devices in wearable smart textiles, encompassing mechanisms, materials, fabrications, device structures, and applications from recent advancements, challenges, and prospects. This review can serve as a valuable reference for researchers in the field of flexible wearable thermoelectric materials and devices and their applications.

Keywords: thermoelectric, weaving, materials, structure, device

1. Introduction

With the accelerated pace of global industrialization, energy consumption has been steadily increasing [1]. Statistics indicate that more than 50% of the thermal energy used in industrial

production dissipates into the atmosphere without re-use [2]. Simultaneously, a substantial amount of energy is expended for cooling buildings and industrial production equipment [3]. Consequently, the effective recovery and utilization of waste heat from industrial processes, as well as the efficient conversion of thermal energy into other forms of energy, have become pressing issues in both the scientific and industrial realms. Against this backdrop, thermoelectric materials and devices, capable of direct conversion between thermal energy and electrical energy, have emerged, showcasing significant potential applications in waste heat recovery and green refrigeration [4–6]. The energy conversion capability

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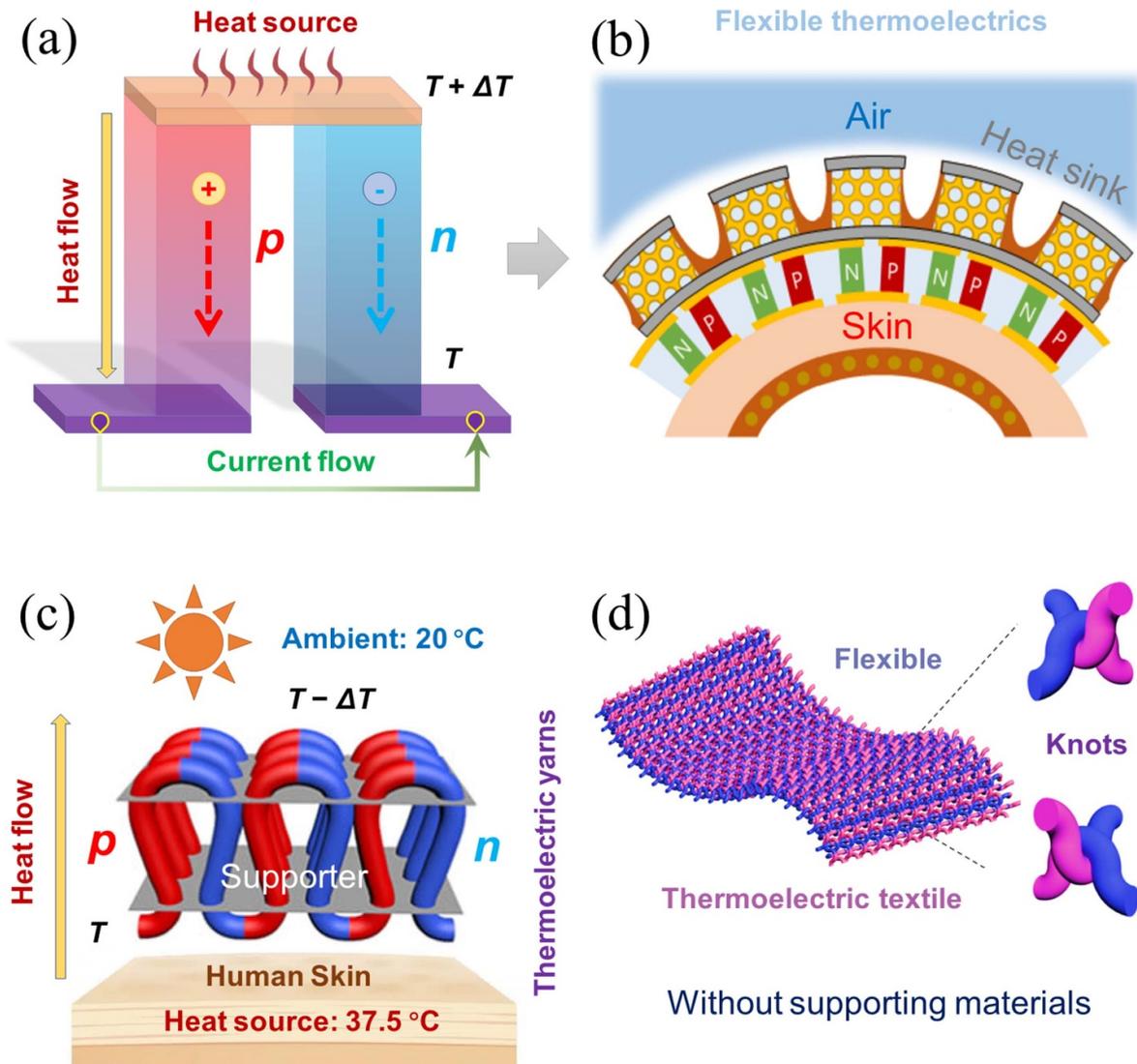


Figure 1. Introduction of weavable thermoelectrics. (a) Illustration of a thermoelectric unit consisting of one pair of n- and p-type bulk thermoelectric materials for power generation driven by a temperature difference (ΔT) between the two sides of the unit. (b) Illustration of flexible thermoelectric units consisting of n- and p-type bulk materials and flexible substrate and heat sinks for power generation by harvesting body heat. Reprinted from [41], © 2019 Elsevier Ltd. All rights reserved. (c) Illustration of weavable thermoelectric units consisting of thermoelectric yarns with both n- and p-type segments for power generation by harvesting body heat. Reproduced from [42]. CC BY 4.0. (d) Illustration of a thermoelectric textile without any supporting materials. Reproduced from [43]. CC BY 4.0.

of thermoelectric materials is quantified by the dimensionless figure-of-merit $ZT = S^2\sigma T/\kappa$, where S is the Seebeck coefficient, σ is the electrical conductivity, and κ is the thermal conductivity of the material, composed of electronic and lattice contributions (κ_e and κ_l), at temperature T [7]. High-performance thermoelectric materials should possess high σ , high S , and low κ . Historically, simultaneously achieving high σ and S to attain a high power factor $S^2\sigma$ is challenging due to the sensitivity of these parameters to the carrier concentration n [8]. Additional structural design is required to reduce the κ since the κ_l is less sensitive to the n [9]. This involves introducing various crystal and lattice defects, such as point defects, dislocations, grain or phase boundaries, pores, and nanoscale inclusions [10]. These defects effectively scatter phonons with different wavelengths, leading to reduced κ_l [11]. However, these defects also scatter carriers to some

extent, leading to decreased carrier mobility μ and in turn, the σ [12]. Therefore, the structural and compositional design of thermoelectric materials has been a persistent challenge.

Thermoelectric devices (TEDs) are typically composed of multiple pairs of n- and p-type thermoelectric materials connected electrically in series and thermally in parallel [13–15]. Figure 1(a) illustrates a pair of thermoelectric materials. Generally, to maximize the thermoelectric conversion efficiency of a device, it is essential to enhance the ZT values of both n- and p-type thermoelectric materials and ensure their compatibility with the device [16–18]. Consequently, researchers have been working diligently to explore new methods to improve the thermoelectric performance of conventional materials [19] and to discover new thermoelectric materials [20]. Computer-assisted approaches, such as first-principles calculations and big data analysis [21, 22],

have been leveraged to expedite these processes. Currently, commercialized thermoelectric materials primarily consist of inorganic bulk materials based on bismuth telluride (Bi_2Te_3), with ZT values around 1 at room temperature and corresponding energy conversion efficiencies of approximately 10% for their device [23]. In contrast, modern household refrigerators achieve energy conversion efficiencies of around 40% using traditional compression-based cooling or steam heat recovery systems [24]. Thus, the energy conversion efficiency of existing TEDs falls short for large-scale industrial waste heat recovery or extensive residential and industrial refrigeration applications. Nevertheless, the advantages of thermoelectric materials and devices are evident [25]. For instance, TEDs are highly stable and easy to maintain. They lack mechanical transmission components or accessories, operate quietly without vibration, and are relatively environmentally friendly, producing no toxic emissions. Additionally, they enable green refrigeration. TED structures are simple and compact, conducive to miniaturization, and adaptable to challenging environments like outer space or remote regions [26]. In fact, over the past few decades, thermoelectric materials have played an indispensable role in certain specialized miniaturized or micro-sized devices. Applications include power generators for space satellites, refrigeration systems for automotive refrigerators, and micro-medical devices, showcasing the versatile potential of thermoelectric materials [3, 13, 23, 27, 28]. Furthermore, through ongoing research, the ZT values of many new thermoelectric materials have already surpassed 2 and can even reach 3 [29], such as GeTe [12, 30, 31], SnSe [32–36], Cu_2Se [37–39]. However, the significant challenge lies in how to effectively apply these new materials in devices. This includes optimizing device structure design based on new materials, refining welding processes, and selecting appropriate transition layers to reduce internal resistance, as well as enhancing mechanical and thermal stability [1, 40]. As a result, the practical application of TEDs based on new materials still faces significant challenges.

With the advancement of technology, there is an increasing demand for multifunctional clothing beyond just warmth, lightness, and aesthetics [44–47]. Wearable functional devices are playing an increasingly vital role in enhancing clothing comfort, improving life quality, enhancing working conditions, and meeting the needs of specific industries and special occasions [48–51]. These wearable devices often incorporate sensors, conductive fibers, nanoparticles, and other intelligent components [52]. However, one of the primary challenges in wearable electronic products is the power supply [53]. Bulky batteries with short lifespans have hindered the practicality of wearable electronics [53]. The use of external power sources limits the scenarios in which wearable electronic products can be used [53]. Attaching batteries to clothing can be cumbersome and heavy, presents capacity issues, and exposes electronic components to the risk of short-circuiting and mechanical failures in contact with sweat or environmental moisture [53]. Therefore, researchers have explored and studied various approaches to optimize traditional textile-based energy sources and search for new flexible power sources [53]. Studies have shown that directly converting environmental

energy into electricity can meet the power requirements of wearable electronic devices [54–58]. Flexible TEDs (F-TEDs), known for their lightweight, compact size, noise-free operation, precision, and reliability, hold significant potential [59–64]. F-TEDs mainly consist of rigid or flexible thermoelectric materials (bulks, films, and fibers), flexible substrates, and sometimes flexible heat sinks [41, 57, 65, 66], as indicated by figure 1(b). In the last decade, the fabrication of wearable electronic devices based on flexible thermoelectric materials has become a new focus in the field of thermoelectric research [56, 58, 61, 66–70]. Flexible thermoelectric materials typically consist of three main categories, namely conductive polymers [71–82], carbon [83–87], and inorganics [88–96]. Conductive polymer is one of the most extensively researched categories of flexible thermoelectric materials [97, 98]. It includes materials like poly(3,4-ethylenedioxythiophene): poly(styrene sulfonate) (PEDOT:PSS) and polyaniline (PANI) with intrinsic low κ due to their polymer nature [24, 77, 98–100]. By adjusting the doping levels of conductive polymers, their σ can be effectively optimized, with the highest σ reaching 10^3 S cm^{-1} [101]. However, these materials typically have relatively low S . Carbon-based materials such as carbon nanotubes (CNTs) and graphene [84, 85], known for their high σ and μ , form a network-like structure when intertwined. This network structure has a low κ , which can reach $0.035 \text{ W m}^{-1} \text{ K}^{-1}$ [102]. However, similar to conductive polymers, carbon materials exhibit relatively low S . Inorganic bulk thermoelectric materials with high σ and S are already in practical use. However, they lack the flexibility to a great extent, although some flexible inorganic films [93, 103, 104] and ductile semiconductors [94, 105–108] have been explored. Besides, nano-structuring inorganic semiconductor materials can effectively enhance their flexibility [67, 109]. By dispersing them in solvents using surfactants, flexible thermoelectric thin films can be prepared [110, 111]. However, surfactants often reduce the σ of inorganic semiconductors to some extent, limiting the effectiveness of this method. To date, the primary means of preparing flexible thermoelectric materials involve chemical synthesis, molecular design strategies, morphology control, and doping techniques to effectively modulate the thermoelectric performance of materials [70, 95].

As a famous branch of flexible thermoelectrics, weavable thermoelectric materials and devices represent one of the forefront research areas. Compared to conventional flexible thermoelectric materials, weavable thermoelectric materials often exhibit higher flexibility and more pronounced one-dimensional (1D) characteristics [54, 89, 96, 112, 113], such as flexible thermoelectric fibers [54, 114–118]. Consequently, they can be woven into flexible substrate materials to create three-dimensional (3D) TEDs using weaving techniques [119]. These flexible substrates can be regular clothing or colloidal or mesh-like support materials, as illustrated in figure 1(c). In some cases, these TEDs can transform into smart textiles without the need for additional flexible substrate support by rational weaving [120, 121], as shown in figure 1(d). Due to the unique structural characteristics of weavable thermoelectric materials and devices, weavable

TEDs (W-TEDs) tend to have higher wearability and possess unique advantages not found in other F-TEDs. For example, W-TEDs can be tailored to match the direction of heat flow between the human body and the environment to maximize the utilization of body heat. W-TEDs also offer notable integration capabilities, a certain degree of washability, compatibility with limb movements, and a wider range of application scenarios. Thermoelectric nanofiber yarns are cost-effective, readily available in terms of raw materials, and possess excellent processing properties. W-TEDs can be easily sewn into fabrics or directly woven into large-area F-TEDs. Simple stitching does not alter the fundamental characteristics of the original fabric, allowing for the incorporation of various electronic functionalities while maintaining comfort and breathability in wearables. Furthermore, their integration into gloves and masks based on thermoelectric effects and the sewability of the yarn can be used for self-powered cold and hot source identification and human respiration monitoring. Self-powered strain sensors composed of yarns can display temperature and voltage changes corresponding to different strains, which can be used to optimize the shooting accuracy of basketball players. These unique features make thermoelectric fiber yarns promising in the field of smart wearables, including wearable generators, respiration monitoring, and sports optimization. As a result, in recent years, weavable thermoelectrics have experienced rapid development. Given the dynamic nature of research in this field, a comprehensive review is urgently needed to carefully summarize the progress within recent years (especially from 2020 to 2023, since there has been no timely review on this topic during this period), and challenges and future trends in this research direction. In this work, we aim to achieve this goal by addressing the aspects mentioned above, with the hope of contributing to the further advancement of research in this direction.

2. Classifications of weavable thermoelectrics

With the advancement of thermoelectric science and technology, the morphology of weavable thermoelectric materials has become diverse, and the research focus in this field continues to evolve. Generally, research focus areas in weavable thermoelectrics can be categorized as indicated by figure 2, including 1D fibers [42, 43, 122–166] and nanowires (NWs) [167, 168], 2D thin films [169–172], 3D fabrics [173–186], device assembling [42, 43, 124], simulations [187, 188], and integrations [189]. Among them, thermoelectric fibers with typical 1D macroscopic morphologies have garnered significant research attention. These fibers include carbon-based materials (such as CNTs) [43, 122–130], organic conducting polymers (such as PEDOT:PSS) [131–147], and some inorganic materials (such as core-shell structured thermoelectric fibers) [148–151]. Composite fibers have also been extensively studied, including composites of carbon and organic materials [42, 152–161], carbon and nano-inorganic materials [162], and composites of organic materials as matrices with nano-inorganic materials as fillers [163–166]. These composite fibers are designed to leverage the unique properties of

different materials, such as the high σ and flexibility of carbon materials, the low κ and high flexibility of organic materials, and the high S of inorganic materials. In some cases, the energy barriers formed at the interfaces between different materials may filter low-energy charge carriers, further enhancing the S without significantly reducing the σ . Additionally, research aimed at improving the mechanical performance (*e.g.* tension and flexibility) and stability of these fibers is essential, as it directly relates to their spinnability, weaveability, and stability. Regarding NWs, their research is currently limited, primarily focusing on process innovation and performance improvement, with little exploration of their spinnability.

As for devices, a key research focus is on effectively weaving optimized thermoelectric fibers into clothing or fabrics. This involves optimizing and innovating textile processes, analyzing device heat flow, and reducing internal resistance. Furthermore, the flexibility, compatibility with human skin, stability, washability, toxicity, and environmental adaptability of woven devices are critical factors to consider. These aspects require extensive experimental validation and computational simulation guidance, often utilizing software such as ANSYS. Another research emphasis includes the design of substrates or supports that match woven thermoelectric fibers, such as common fabrics, flexible strings, and silicone. These support materials can serve solely as structural supports [173–175] or be thermoelectrically functionalized [176–186].

Lastly, to achieve the goal of smart textiles, woven devices based on thermoelectric fibers are typically not used in isolation but in conjunction with other functional devices. These may include wearable current-voltage regulators (*e.g.* amplifiers), energy storage units (*e.g.* miniature supercapacitors), and other functional devices such as triboelectric generators, moisture generators, and more, to achieve complex objectives like electronic skin and personal thermal management. In the following chapters, we will discuss the latest developments and challenges in each of these research focus areas.

3. Configurations of weavable thermoelectrics

The configurations of W-TEDs have various impacts on their performance [113]. Generally, these factors include material selection, structural design, heat dissipation system, size and shape, temperature gradient, and connection and wiring. Firstly, the main performance of W-TEDs is closely linked to the selection of weavable materials. The ZT s of weavable thermoelectric materials are critical parameters that affect the thermoelectric efficiency of their devices. Large ZT values usually lead to high thermoelectric efficiencies. Also, the flexibility and wearability of the materials significantly influence the flexibility and wearability of their devices. Secondly, based on high-performance weavable thermoelectric materials, the structural design of devices can also affect their thermoelectric performance. Typically, W-TEDs consist of multiple thermoelectric elements connected through electrodes. The design of the structure needs to consider how to minimize thermal losses while maximizing the transmission of electric current. Thirdly, W-TEDs generate heat during

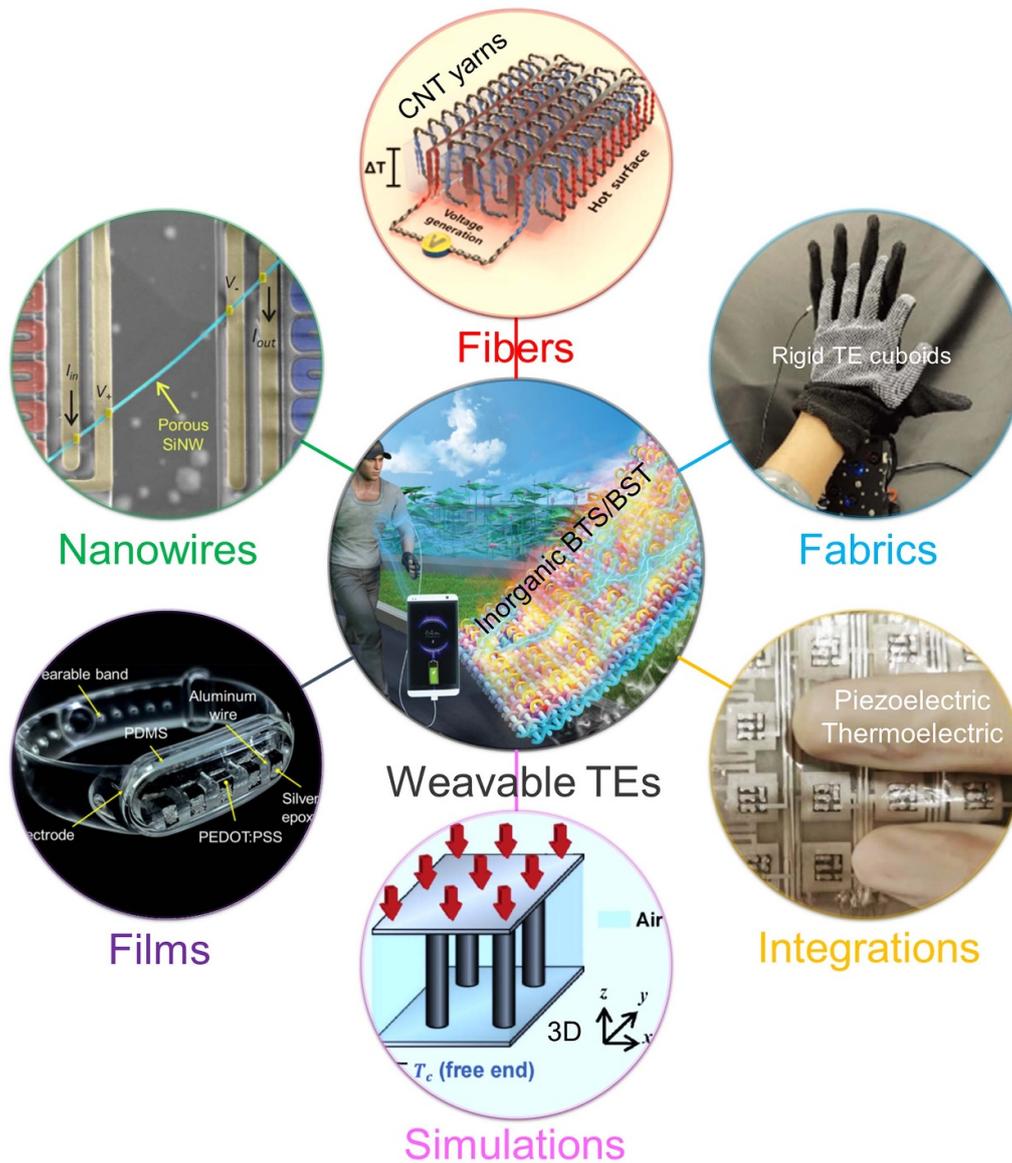


Figure 2. Classifications of weavable thermoelectrics. In the center: illustration of employing weavable thermoelectrics (TEs) for continuous charging of a smartphone by harvesting human body heat. Here BTS and BST are n-type $\text{Bi}_2\text{Te}_{3.3}\text{Se}_{0.2}$ and p-type $\text{Bi}_{0.4}\text{Sb}_{1.3}\text{Te}_3$, respectively. Reproduced from [150] with permission from the Royal Society of Chemistry. Fibers: n/p-type carbon nanotube (CNT) yarns. [123] John Wiley & Sons. © 2022 Wiley-VCH GmbH. Fabrics: composed of rigid TE cuboids (p-type: $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ /n-type: $\text{Bi}_2\text{Te}_{2.7}\text{Se}_{0.3}$). Reproduced from [173]. CC BY 4.0. Integrations: piezoelectric poly(vinylidene fluoride-co-trifluoroethylene) and thermoelectric polyaniline-based composites. [189] John Wiley & Sons. © 2020 Wiley-VCH GmbH. Simulations: models of three-dimensional (3D) array generator with a low temperature difference under conductive and radiative heat transfer. Reprinted from [188], © 2020 Elsevier Ltd. All rights reserved. Films: vertically aligned p-type poly(3,4-ethylene-dioxythiophene):poly(styrene sulfonate) (PEDOT:PSS) thin film and n-type aluminum wire-based thermoelements integrated wearable device. Reprinted from [172], © 2022 Published by Elsevier Ltd. Nanowires: silicon nanowire (NW) under thermoelectric performance evaluation. Reproduced from [167]. CC BY 4.0.

operation and may require an effective heat dissipation system to maintain the proper temperature. The heat dissipation structure and materials of the device also influence its performance. Improved heat dissipation can enhance thermoelectric efficiency. Fourthly, the size and shape of W-TEDs also impact their performance. Generally, larger devices can generate more electrical energy but also require more thermal input. Therefore, design considerations must balance size and performance requirements. The performance of W-TEDs relies on thermoelectric effects, meaning that they require the presence

of a temperature gradient (one side hotter, one side cooler) to operate. Thus, structural design must ensure an appropriate temperature gradient exists. Finally, the design of electrode connections and wiring can also affect performance. Electrodes should effectively collect and transmit current to maximize the efficiency of energy conversion.

Although there have been reports on combining traditional textiles with thermoelectric technology to produce weavable thermoelectrics, related research is still in its initial exploration stage. Existing weavable thermoelectrics have relatively low

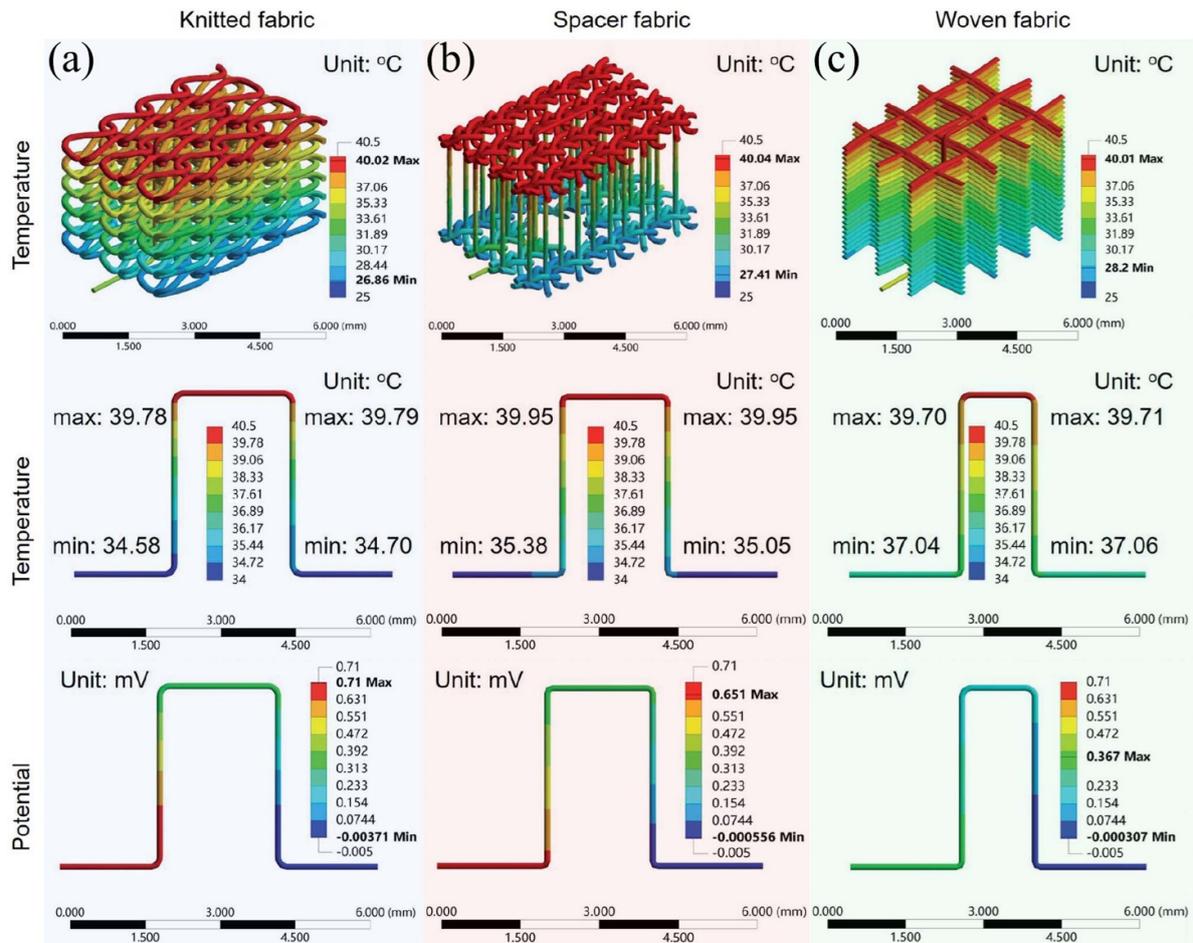


Figure 3. Configurations of weavable thermoelectrics. Finite element simulations of three types of thermoelectric fabrics including (a) knitted, (b) spacer, and (c) woven fabrics. Reproduced from [124] with permission from the Royal Society of Chemistry.

electrical power output, poor wearability, and low resistance to deformation, and research on the influence of fabric structure on thermoelectric output performance is still a blank area in the field. The design concept of W-TEDs aims to utilize the temperature difference ΔT between the human body and the environment to achieve self-powered wearable electronic devices while ensuring comfort. Therefore, some efforts have been made to analyze the influence of device structure on the coupling effects of temperature and electric potential fields using multi-physics finite element simulations, providing theoretical and technical support for the subsequent development of high-performance weavable thermoelectrics. Figure 3 compares finite element simulations of three types of thermoelectric fabric-based modules including knitted, spacer, and woven fabrics by ANSYS software [124]. In this figure, the first row displays different weaving methods, while the second and third rows represent the corresponding temperature distribution and electric potential distribution, respectively. Here, a fixed heating temperature is applied to the top surfaces (hot sides) of the modules, and the bottom surfaces (cold sides) of the modules are exposed to ambient air through thermal convection. At low temperatures, heat transfer within the module is primarily through convection and conduction, making thermal radiation negligible. Taking a hot-side temperature

of 40 °C as an example, different weaving methods result in varying thermal resistance within the module, consequently leading to changes in temperature distribution and the corresponding electric potential distribution within the device. The results show that the knitted fabric structure exhibits the largest ΔT , indicating the highest thermal resistance, thereby absorbing the least amount of heat, resulting in the maximum effective temperature gradient within the thermoelectric legs and the highest achievable voltage and anticipated output power. Additionally, woven fabrics typically have regular yarn arrangements in the thickness direction, reducing the number of closed channels that can trap static air to suppress thermal convection in woven fabrics. While knitting loops in the knitting structure are periodically interconnected, woven fabrics such as weft-knitted fabric or warp-knitted spacer fabric generally have irregular yarn arrangements overall, allowing them to trap more static air to reduce thermal convection within these fabrics. Therefore, fabric structure can significantly impact the power generation of the device, and careful selection and design of the fabric structure are crucial for maximizing the ultimate performance of the device. Other research has also found that, under well-designed weaving processes, the additional use of encapsulation techniques can effectively prevent short circuits in thermoelectric modules.

Finite element analysis and practical test results have shown that insulating encapsulation layers can increase the ΔT at both sides of the thermoelectric legs and the heat flow passing through the thermoelectric legs, thereby enhancing output performance [42].

To better keep a high ΔT between the hot and cold sides of the device, some works report that for W-TEDs, a 3D device architecture is superior to a 2D device architecture [42]. Generally, achieving an effective architecture aligned with the out-of-plane heat flow direction poses a primary challenge. To meet this requirement, a 3D architecture can be formed by interlocking thermoelectric yarn loops. The interlocking pattern allows these loops to automatically stand at an angle along the out-of-plane direction due to the elastic force of bending fibers, eliminating the need for a substrate. This 3D textile is characterized by rows of interlocked loops, and the angle is determined by weaving parameters. Notably, this interlock mode is applicable to large-area fabrics in the textile industry. The resulting thermoelectric modules are compact and effective, with p/n legs electrically connected in series and thermally in parallel. In contrast, a conventional configuration where the pillar position of one thermoelectric loop is covered by the arc position of the next loop, resulting in a 2D architecture. 3D architecture exposes electrodes on one surface for improved thermal contact, while the thermoelectric legs wrapped with fibers lie flat on the other surface. The comparison of the output voltages reveals that the 3D architecture offers approximately 24 times higher output voltage compared to the 2D configuration. By changing the interlock mode, a conventional 2D device can be transformed into a high-performance 3D architecture. Besides, it was reported that directly woven 3D thermoelectric units exhibit superior ΔT and voltage compared to fibers alone through a 3D textile substrate [42]. This approach, based on elastic force rather than a supporting substrate, differentiates it from other textile-based W-TEDs, offering improved mechanical properties, stability, and output power.

4. Carbon-based weavable thermoelectrics

In W-TEDs, in addition to optimizing the structure of the device (such as the influence of weaving methods discussed in the previous section), the selection and performance optimization of weavable thermoelectric materials are of paramount importance. Among these materials, 1D carbon fibers, such as single-walled, double-walled, or multi-walled CNTs (SW/DW/MWCNTs), typically exhibit high σ (generally $>1000 \text{ S cm}^{-1}$) [84]. This is crucial for thermoelectric applications, as high σ enables better electrical current conduction, thereby improving thermoelectric efficiency. Although carbon fibers tend to have lower S (usually $<30 \mu\text{V K}^{-1}$) [84], their S values can be optimized to some extent by surface modification of the carbon fibers. This can even lead to the p-n transformation of carbon fibers. Also, carbon fibers often possess high κ due to the nature of carbon [84], which is detrimental to thermoelectric performance. Therefore, to reduce κ , the structure of carbon fibers needs to be carefully designed. For

instance, designing carbon fibers with a mesh-like structure and a certain degree of porosity is an effective way to lower κ .

In addition to their tunable thermoelectric properties, 1D carbon fibers offer several other advantages as materials for weavable thermoelectrics. For instance, carbon fibers are lightweight yet exceptionally strong materials. They have a high specific strength, meaning they can withstand substantial mechanical stress while remaining relatively lightweight. This is highly advantageous for creating lightweight and durable TEDs. Carbon fibers typically exhibit good chemical stability, with the ability to endure high temperatures and chemical corrosion. This makes them capable of maintaining stable performance in various environmental conditions. Carbon fibers can be fabricated into different shapes and sizes, allowing them to adapt to a wide range of TED designs. Consequently, much recent research has focused on using carbon fibers in the design of W-TEDs.

In recent years, the mostly focused carbon-based weavable thermoelectric materials are p- and n-type CNT fibers or yarns [122–130]. To illustrate the progress of carbon-based weavable thermoelectrics, figure 4(a) shows the fabrication of SWCNT-based yarns with n- and p-type segments. The flexible thermoelectric yarns composed of SWCNTs and polyvinyl alcohol (PVA) hydrogels were prepared using a continuous alternating extrusion process [43]. The rheological properties of the gel after solidification showed significantly higher shear storage moduli G' and loss moduli G'' values, with an amplification of approximately three orders of magnitude [43]. In addition to the various tunabilities of the hydrogel, another notable advantage is the limitation on solvent migration due to the PVA polymer network [43]. Modulating the ratio of polyethyleneimine (PEI) to SWCNTs can determine the major charge carriers in the segments, thereby easily forming p- or n-type segments. The electrical resistance of the composite material increases with an increase in PEI content, and the high S aids in temperature gradient sensing [43]. A balanced fiber voltage and current were achieved with a 25% ratio. Both the voltage and current of the fiber linearly increase with the ΔT [43]. Figure 4(b) illustrates the structures of the n- and p-type segments and the interface between them. The gel produced by alternating extrusion ensures that the p/n interface has clear boundaries even under continuous axial compressive and shear stresses [43]. This technique can be used to fabricate mechanically robust and flexible carbon-based thermoelectric fibers, and it offers a simple, controllable, and industrially scalable process [43]. Leveraging the advantages of the hydrophilic colloidal structural network and its rheological properties, the superior confinement of non-uniform particles within the continuous matrix results in highly uniform, well-bonded alternating p/n segments. This continuous arrangement of p/n-type fibers greatly reduces the complexity of textile electronics integration and opens up possibilities for multifunctional devices, including non-planar energy harvesting and multi-pixel touch panels for writing and communication [43]. Figure 4(c) illustrates a photo of a woven textile that is washable. Through appropriate encapsulation processes, it is possible to achieve waterproofing for both the fibers and

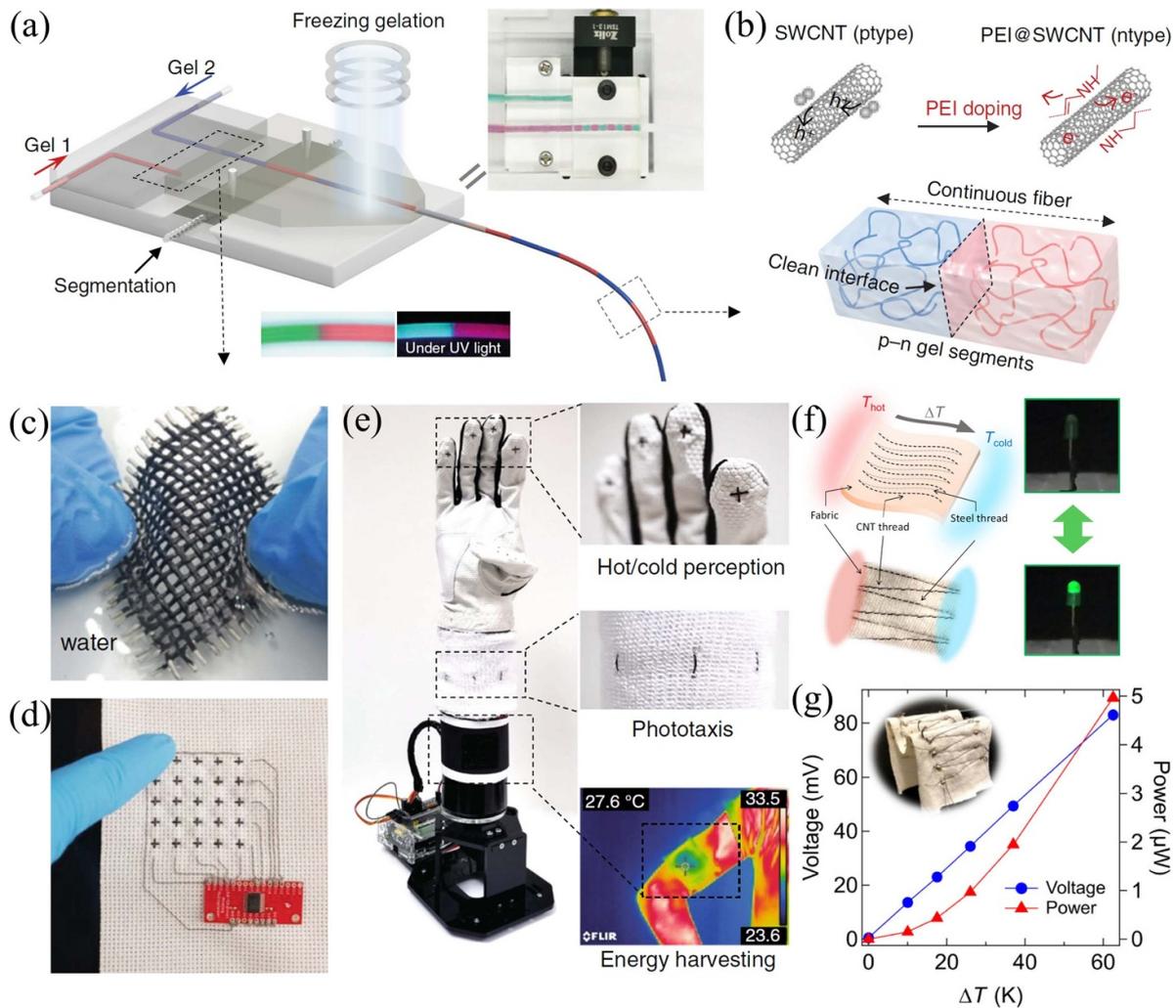


Figure 4. Carbon-based weavable thermoelectrics. (a) Illustration of fabricating single-walled carbon nanotube (SWCNT) based yarns with n- and p-type segments by a continuously alternating extrusion process. (b) Illustration of the structures of the n- and p-type segments and the interface between them. Here PEI is abbreviated from polyethyleneimine. (c) Photo of cross-stitched yarns to form a touch panel. (d) Photo of cross-stitched yarns to form a touch panel. (e) Photo of a robotic arm wearing multifunctional textiles. Reproduced from [43]. CC BY 4.0. (f) Illustration of a weavable thermoelectric device (W-TED) composed of CNTs for power generation. (g) Measured ΔT -dependent voltage and power for the foldable device shown as an inset. Reproduced from [129]. CC BY 4.0.

the devices woven from them [43]. Also, the fiber exhibits excellent mechanical properties and can withstand high tensile strengths of over 20 MPa, whether doped with PEI or not [43]. Moreover, the binding between adjacent p/n segments is strong, capable of withstanding a weight of 500 g [43]. Even when bent into various shapes, the fiber maintains satisfactory electrical performance.

Utilizing the good mechanical properties of thermoelectric fibers with continuous p/n units, it is easy to weave 10 of these fibers into a cross-stitch pattern, forming thermoelectric textiles for touchscreen and communication applications. Figure 4(d) shows a photo of cross-stitched yarns to form a touch panel [43]. When a specific node comes into contact with a heat source, the temperature at the contact point is much higher than that of adjacent nodes. The voltage signals correspond closely and linearly to the temperature of the object, laying the foundation for touch positioning. Further input of handwritten letters was achieved through a

5×5 pixel touch panel. Due to the superior light-absorbing ability of carbon materials, the fibers easily heat up under illumination. The photothermal effect generates a temperature gradient between the illuminated and non-illuminated regions, thereby producing electrical signals. Leveraging the photothermal sensing capabilities, thermoelectric optical fibers were constructed on all six faces of a cube, successfully achieving optical communication, and accurately sensing the direction of incident light. Figure 4(e) shows a photo of a robotic arm wearing multifunctional textiles [43]. Robotic arms wear modular thermoelectric clothing to perform multiple tasks. The robotic arms are equipped with thermoelectric fiber gloves, wristbands, and sleeves, which provide thermal/cold sensing capabilities, photosensitivity, and energy harvesting capabilities. This textile designed for collecting human body heat can also be scaled up and integrated into other wearable self-powering systems, allowing for battery charging or direct power supply to electronic devices. With this set of thermoelectric textiles,

robots can engage in multitasking, not limited to sensing and energy harvesting.

Other works also reported carbon-based weavable thermoelectric materials and their devices, and most of these works primarily focus on improving the thermoelectric performance of carbon fibers themselves. Figure 4(f) illustrates a W-TED composed of CNTs for power generation [129]. The macroscopic woven CNT fibers exhibit exceptionally high σ and in turn a remarkably high $S^2\sigma$ of $14 \pm 5 \text{ mW m}^{-1} \text{ K}^{-2}$ [129]. The high $S^2\sigma$ is related to the adjustment of the inherent Fermi level of the nanotubes, which determines the electrochemical potential. Researchers can control the Fermi level through chemical doping, allowing them to tune the electronic properties of the fibers. Figure 4(g) shows measured ΔT -dependent voltage and power for the foldable device shown as an inset [129]. An output power P of $5 \mu\text{W}$ can be achieved when a ΔT of 60 K is applied to the device.

However, despite the clear advantages of carbon materials as the core of weavable thermoelectric materials and devices, their drawbacks are also quite evident. Compared to some other thermoelectric materials, carbon-based devices exhibit relatively lower thermoelectric efficiency [190, 191]. While they possess high σ , their κ is also relatively high, which can result in reduced thermoelectric performance. Carbon fibers typically have lower S , limiting their efficiency in certain thermoelectric applications. The production of high-quality carbon fibers often involves expensive processes, including carbonization and textile manufacturing, which can increase material costs. The thermoelectric performance of carbon fibers may be limited to extremely high or low temperatures, restricting their use in some high-temperature or low-temperature applications. In some cases, carbon fibers may exhibit brittleness and lack sufficient flexibility or stretchability, which can limit their use, especially in applications requiring bending or twisting. With the advancement of science and technology, researchers may find ways to improve the thermoelectric performance of carbon fibers or develop alternative materials more suitable for specific applications.

5. Inorganic weavable thermoelectrics

Currently, the vast majority of thermoelectric materials with practical utility are inorganic semiconductor thermoelectric materials [1]. This is because such materials possess relatively high ZT s derived from their high σ , S , and low κ [192, 193]. Furthermore, the performance of inorganic materials can be adjusted and optimized through chemical synthesis and process control to meet specific application requirements. Many inorganic materials also exhibit outstanding high-temperature stability [1], making them suitable for high-temperature thermoelectric applications. Inorganic materials typically have superior mechanical strength and durability, making them suitable for environments that require resistance to mechanical stress or vibration. They often exhibit good chemical stability, resisting corrosion and oxidation, thereby extending their lifespan. Some inorganic materials are also abundant in resources and can be obtained through sustainable mining and

production methods. Therefore, researchers often aim to translate these advantages into W-TEDs.

To prepare 1D inorganic thermoelectric fibers while maintaining their high thermoelectric performance, the molten core process is a promising approach. Through thermal drawing, inorganic materials can be transformed into fine and elongated fibers that typically exhibit good flexibility and bending properties, making them suitable for F-TEDs. Additionally, the thermal drawing process allows for uniform stretching and size control of materials, resulting in fibers with improved uniformity. This preparation method often boasts high industrial scalability, enabling the production of large quantities of consistently high-quality thermoelectric fibers. Figure 5(a) illustrates the fabrication of flexible $\text{Ag}_2\text{Te}_{0.6}\text{S}_{0.4}$ fiber by a thermal drawing process [148]. This process can produce core-shell-like fiber bundles with relatively homogeneous structure and composition, achieving a $S^2\sigma$ of $\sim 600 \mu\text{W m}^{-1} \text{ K}^{-2}$. Furthermore, these fibers can withstand a tensile strain of 21.2%, allowing them to undergo various complex deformations. Importantly, the thermoelectric performance of the fibers exhibits high stability even after approximately 1000 cycles of bending and releasing processes, with a bending radius of up to 5 millimeters. Figure 5(b) illustrates the structure of the W-TED composed of $\text{Ag}_2\text{Te}_{0.6}\text{S}_{0.4}$ fibers supported by polyester fabric. The high flexibility of the inorganic fibers allows for their integration into 3D wearable textiles. Figure 5(c) illustrates wearing the as-fabricated device for power generation by harvesting the body heat [148], which results in a standardized power density of $0.4 \mu\text{W m}^{-1} \text{ K}^{-2}$ at a ΔT of 20 K. This is comparable to high-performance Bi_2Te_3 -based inorganic thermoelectric fabrics [194] and is nearly two orders of magnitude higher than carbon-based and organic thermoelectric fabrics [114, 115]. It should be noted that the process of using thermally drawn inorganic materials to manufacture weavable thermoelectric materials and devices also faces some challenges, including material selection, process control, equipment, and other issues. Choosing the right inorganic materials and maintaining their high thermoelectric performance in the manufacturing process requires careful material design and engineering control. Additionally, the investment costs for equipment and processes may be high, requiring specialized knowledge and skills for implementation.

In addition to the thermal drawing process to achieve high flexibility, it is also possible to mimic the n/p segments of carbon fibers discussed above to prepare chain-like inorganic thermoelectric wires with n/p segments for assembling weavable devices. Figure 5(d) illustrates assembling thermoelectric chains composed of n- and p-type inorganic segments [150]. The manufacturing process of this thermoelectric chain can be simplified into the following steps. Firstly, polyimide (PI) filaments are embedded as a complete sequence into holes of a graphite mold. Then, p- and n-type thermoelectric powders are alternately placed into the mold, and under a pressure of 200 MPa with the assistance of a tungsten-steel frame outside the graphite mold, they are pressed for 1 h. Subsequently, the bead-like chain is demolded and sintered through ultrafast high-temperature sintering processing with a current of 5.5 A and applied power of 91 W to obtain the

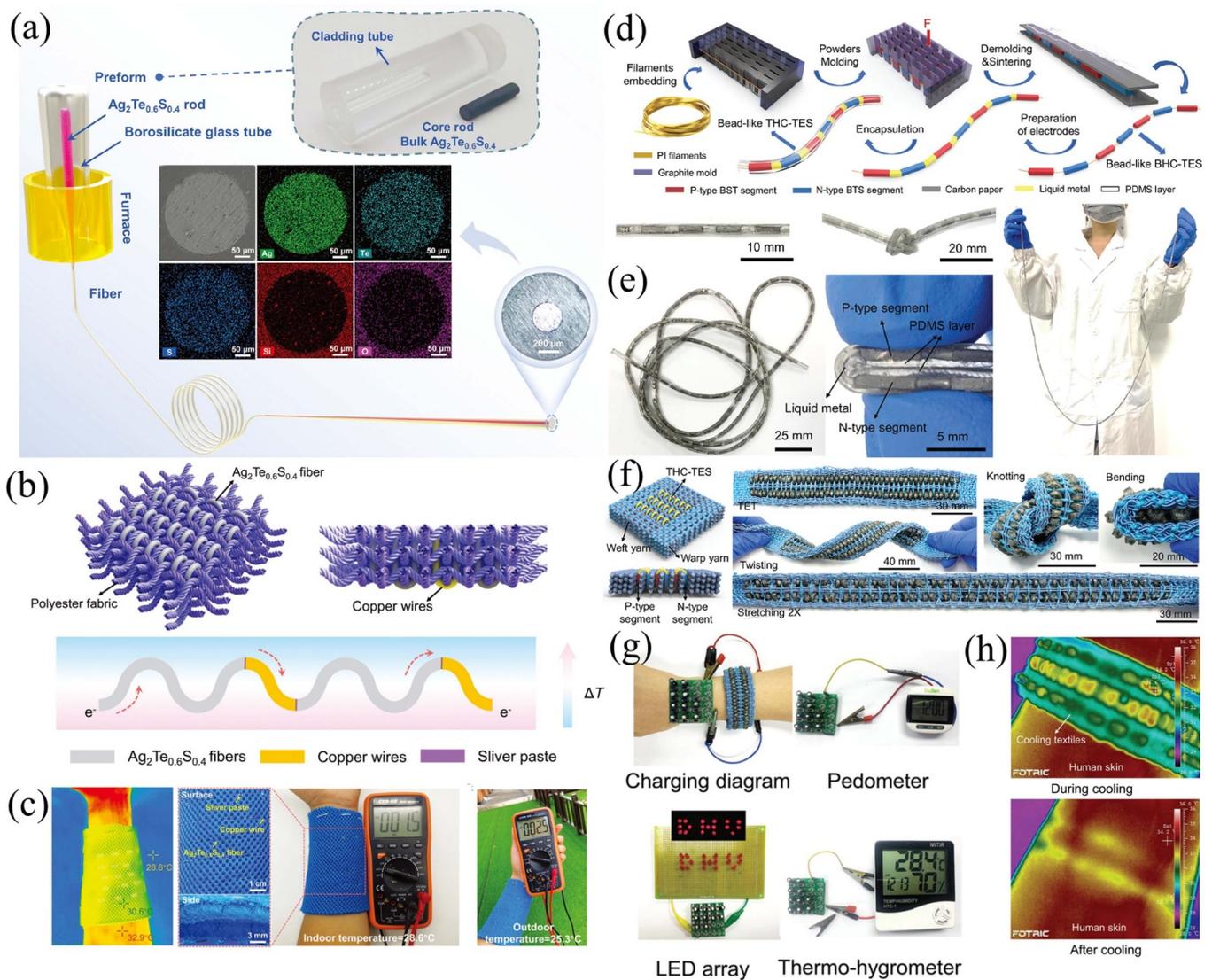


Figure 5. Inorganic weavable thermoelectrics. (a) Illustration of fabricating flexible $\text{Ag}_2\text{Te}_{0.6}\text{S}_{0.4}$ fiber by a thermal drawing process. (b) Illustration of the structure of the W-TED composed of $\text{Ag}_2\text{Te}_{0.6}\text{S}_{0.4}$ fibers supported by polyester fabric. (c) Illustration of wearing the as-fabricated device for power generation by harvesting body heat. Reproduced from [148]. CC BY 4.0. (d) Illustration of assembling thermoelectric chains composed of n- and p-type inorganic segments. (e) Photos of the as-fabricated chains. (f) Illustration of weaving the chains in a fabric. (g) Photos illustrating the as-fabricated wearable device for power generation. (h) Infrared images exhibiting the cooling effect of the wearable device. Reproduced from [150] with permission from the Royal Society of Chemistry.

formed thermoelectric chain. Each p/n segment in the thermoelectric chain is 5 millimeters long. Then, $\text{Ga}_{75}\text{In}_{25}$ liquid metal is used as an interconnection electrode between the p- and n-type thermoelectric segments. The length of each liquid metal electrode is 2 millimeters. Finally, the strings are wrapped and encapsulated with a polydimethylsiloxane (PDMS) layer to form the finished product. The thickness of the PDMS layer is 0.5 millimeters. Figure 5(e) shows photos of the as-fabricated chains. The thermoelectric chain can be as long as 1.2 m and exhibits excellent flexibility. The bead-like thermoelectric chain can be knotted and bent with a bending radius of 2 millimeters and a bending angle of 180 degrees. Figure 5(f) illustrates the chains in a fabric. By weaving these thermoelectric chains into the fabric substrate, these optical images demonstrate their flexibility and

stretchability, including twisting, knotting, and bending. The bending radius is 2 mm, and they can stretch up to 100%. It also exhibits certain washability confirmed by ~ 20 washing cycles. Figure 5(g) illustrates the as-fabricated wearable device for power generation [150]. At a ΔT of 25 K, the device output power density is 0.58 W m^{-2} . The predicted power density at a ΔT of 80 K is 6.06 W m^{-2} . Finite element analysis has demonstrated the significance of the fabric structure in achieving its excellent mechanical and thermoelectric performance, achieving an output voltage of 0.28 V at an environmental temperature of 8° . Figure 5(h) shows infrared images exhibiting the cooling effect of the wearable device [150]. The device can stably produce a solid-state cooling effect of 3.1 K in static air at an environmental temperature of 26°C and a relative humidity of 60%. It can also continuously power

wearable electronic devices when worn on a self-generated arm temperature gradient of 16 K, used for monitoring environmental and human life signals and activities.

However, the reality is that applying inorganic materials to W-TEDs is highly challenging. Firstly, many inorganic thermoelectric materials are prone to fracture or breakage in flexible applications because they are typically brittle. F-TEDs need to withstand deformations such as bending, stretching, and twisting, but these materials often lack the necessary flexibility. Secondly, the mechanical properties of inorganic thermoelectric materials are typically insufficient to meet the high bending performance required for F-TEDs. In flexible applications, materials need to undergo multiple bending cycles without damage. Additionally, some inorganic thermoelectric materials are relatively heavy, which can be problematic for flexible applications, as lightweight properties are crucial for the comfort and portability of flexible materials. Moreover, the complex preparation processes of some inorganic thermoelectric materials make it challenging to achieve uniform coating or processing on flexible substrates, increasing the manufacturing difficulty and cost of flexible devices. Ensuring that inorganic materials can maintain high performance in bulk form after being made flexible is also a challenge. Finally, the high-performance characteristics of certain inorganic thermoelectric materials may be optimized for high-temperature ranges, whereas W-TEDs may need to operate at or near room temperature, which limits the full utilization of their high performance. Therefore, it is of great significance to develop flexible, non-toxic, and high- ZT inorganic thermoelectric materials for near-room-temperature wearable applications, and innovation in the fabrication processes is also an important goal.

6. Organic weavable thermoelectrics

Organic materials as thermoelectric materials have several advantages, especially in specific applications such as flexible and wearable textile-based electronic devices. Organic materials are typically more flexible and easily bendable compared to inorganic materials. This makes them highly suitable for manufacturing F-TEDs that can conform to various shapes and curved surfaces. They are also lighter than inorganic materials, which is crucial for the comfort and portability of flexible and wearable devices. Also, organic materials often have the potential for low-cost fabrication because they can be processed using techniques such as printing, spraying, and other solution-based methods without the need for high-temperature processing or expensive equipment [67, 71]. Their properties can be tailored and optimized for specific applications through molecular design and synthesis, providing flexibility and customization as needed. Besides, organic materials are typically more biocompatible, making them suitable for applications involving contact with biological systems, such as medical devices or biosensors. In comparison to some inorganic thermoelectric materials, organic materials often operate at lower temperatures, which is advantageous for certain low-temperature applications. Last but not least,

many organic materials can be obtained through sustainable production methods, reducing reliance on hazardous chemicals and scarce resources. The electrical properties of organic materials, such as σ and S , can often be adjusted through molecular design and chemical modification to optimize thermoelectric performance [195, 196].

Generally, PEDOT:PSS fiber is one of the most focused organic weavable thermoelectric materials [131, 136, 142, 144, 145], which is usually fabricated by wet-spinning technique [133, 134, 137, 138, 140, 143, 146]. Sometimes organic composites are designed to meet specific requirements, such as PEDOT:PSS/PVA [135, 139, 197], PEDOT:PSS/waterborne polyurethane (WPU) [132], PEDOT:PSS/poly(ethylene glycol) (PEG) [147], *etc.* In addition to the traditional wet-spinning method, one of the most advanced techniques to fabricate organic fibers for assembling weavable thermoelectrics is 3D printing. 3D printing technology allows for easy customization and design adjustments as needed. This makes it possible to manufacture organic material products with specific shapes, sizes, and properties to meet the requirements of various applications. Particularly, it enables the creation of complex geometric shapes, including internal structures and hollow parts, which can be challenging or impossible to achieve using traditional manufacturing methods. Additionally, 3D printing is an additive manufacturing process that can reduce waste and material waste. This contributes to cost reduction and improved sustainability. It enables rapid prototyping, shortening the product development cycle, and allows for flexible batch customization to meet individualized demands. In some cases, 3D printing of organic materials can lead to reduced production costs. Figures 6(a)–(c) illustrate the composition of PEDOT:PSS solution, printable PEDOT:PSS ink, and 3D-printed PEDOT:PSS products, respectively [141]. By freeze-drying and redispersing the original PEDOT:PSS solution under low-temperature conditions, it can be transformed into a printable conductive polymer ink. Through dry annealing and subsequent swelling in a wet environment, the 3D-printed conductive polymer can be converted into both a dry state and a hydrogel state of pure PEDOT:PSS. Figure 6(d) illustrates the photo of a 3D-printed PEDOT:PSS mesh after dry-annealing. PEDOT:PSS can be easily manufactured into high-resolution and high aspect ratio microstructures, and can be integrated with other materials such as insulating elastomers through multi-material 3D printing. The 3D-printed PEDOT:PSS can also be transformed into highly conductive and flexible hydrogel microstructures, as indicated by figure 6(e) [141]. Figure 6(f) also shows a photo of the foldable 3D-printed PEDOT:PSS circuit patterns [141]. These results indicate that this process enables the rapid and simplified manufacturing of various conductive polymer devices.

In addition to advanced manufacturing process innovations, the pursuit of higher thermoelectric performance and good flexibility and stability has always been a focus for organic weavable thermoelectric fibers and devices. Figure 6(g) illustrates the fabrication of PEDOT:PSS fibers by a conventional wet-spinning process [134]. By adjusting the number of freeze-thaw cycles, extrusion speed, needle length-to-diameter ratio, and the type of coagulation

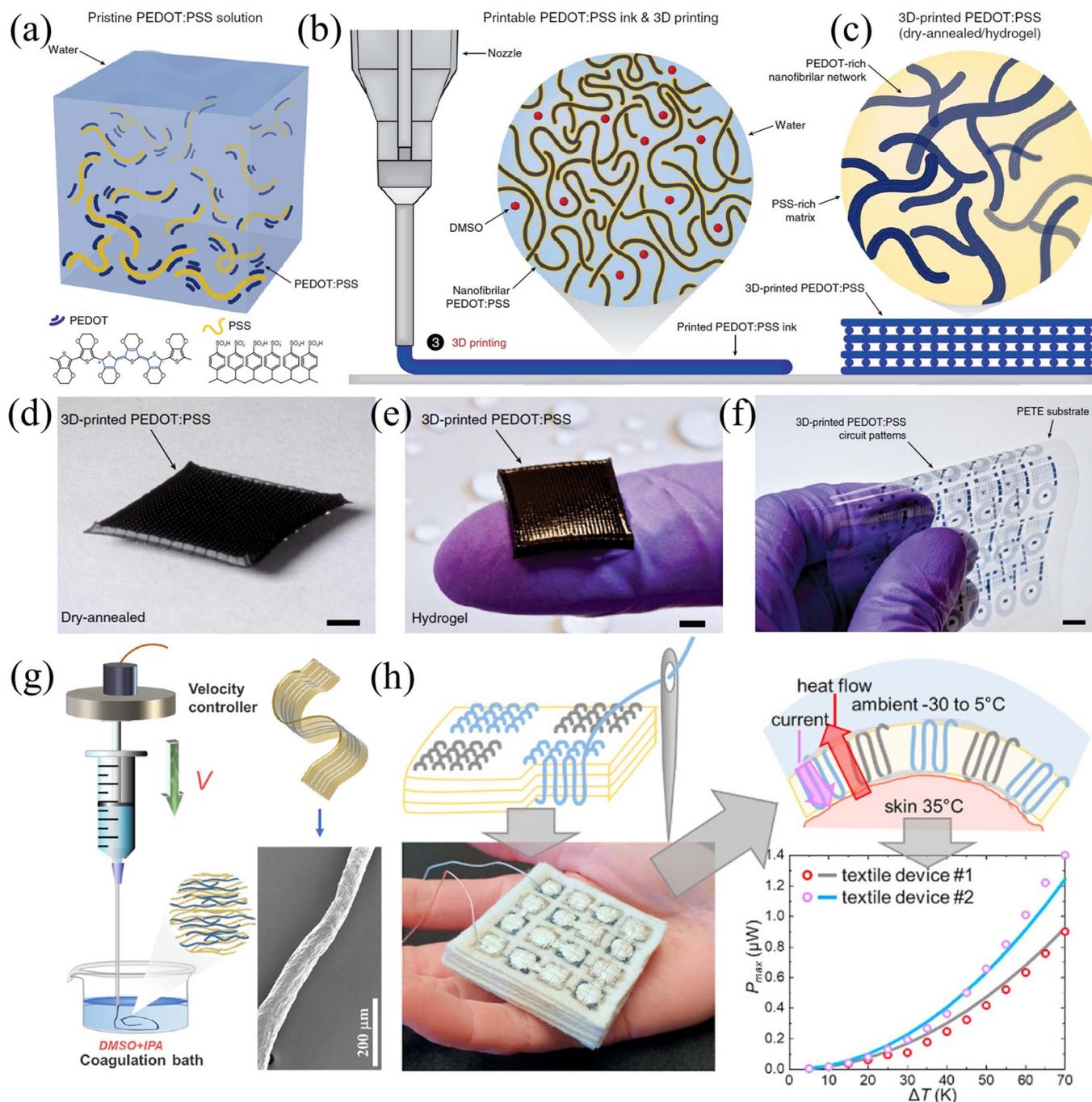


Figure 6. Organic weavable thermoelectrics. (a) Illustration of the composition of PEDOT:PSS solution. (b) Illustration of printable PEDOT:PSS ink. (c) Illustration of three-dimensional (3D) printed PEDOT:PSS. (d) Photo of a 3D-printed PEDOT:PSS mesh. (e) Photo of the mesh in a hydrogel state. (f) Photo of the foldable 3D-printed PEDOT:PSS circuit patterns. Reproduced from [141]. CC BY 4.0. (g) Illustration of fabricating PEDOT:PSS fibers by a wet-spinning process. Reprinted from [134], © 2022 Elsevier Ltd. All rights reserved. (h) Illustration of weavable PEDOT:PSS yarns and the performance of the woven device. Reproduced from [136]. CC BY 4.0.

bath, the spinnability and thermoelectric performance of the PEDOT:PSS spinning solution were significantly improved. Therefore, optimized PEDOT:PSS fibers obtained at relatively high extrusion speeds (150 mm min^{-1} or 1037 ml h^{-1}) exhibited a remarkably high σ of $1013 \pm 32 \text{ S cm}^{-1}$ and excellent mechanical stability under large mechanical bending or twisting deformations. Based on PEDOT:PSS fibers, a simple and flexible generator was further assembled, capable

of generating stable output performance by collecting human body heat or low-temperature ice, demonstrating its adaptability to various scenarios. For the weavability evaluation, figure 6(h) illustrates weavable PEDOT:PSS yarns and the performance of the woven device [136]. The conductive sewing yarns were embroidered into thick wool fabric to create a planar out-of-plane thermoelectric textile generator. Using models developed for traditional thermoelectric systems, the

performance of electronic textile devices can be accurately predicted and optimized as long as the model includes electrical contact resistance and thermal contact resistance. The thermoelectric textile device generated $1.2 \mu\text{W}$ of power at a ΔT of 65 K, and over $0.2 \mu\text{W}$ of power at a more moderate ΔT of 30 K.

It should be noted that while organic materials based on conductive polymers have excellent weavability, they also face significant disadvantages and challenges. In comparison to some inorganic materials, organic materials typically exhibit lower σ and S , limiting their thermoelectric performance [198]. This means their devices generate relatively less electrical energy under the same ΔT . Additionally, many organic materials are susceptible to decomposition or degradation at high temperatures or over prolonged usage, reducing their stability and operational lifespan. Furthermore, organic materials often possess lower mechanical strength, making them prone to breakage or wear, limiting their durability in certain applications. Moreover, organic materials tend to be more sensitive to environmental factors such as humidity, oxygen, and UV radiation, which can lead to performance degradation or failure. While the production of organic thermoelectric materials is generally more cost-effective than some inorganic counterparts, achieving high-performance organic thermoelectric materials may still require precise synthesis and processing steps, potentially increasing costs. Lastly, most organic materials exhibit good thermoelectric performance within a relatively low-temperature range, with performance diminishing at extremely high temperatures, restricting their utility in high-temperature applications. However, while organic materials have these drawbacks, they still have advantages in certain specific applications, especially in areas requiring flexibility and lightweight characteristics, such as wearable electronic devices and textile electronics. Furthermore, researchers are continually working to enhance the performance of organic thermoelectric materials to address these challenges.

7. Carbon/inorganic hybrid-based weavable thermoelectrics

By combining carbon materials with traditional inorganic thermoelectric materials, it is possible to create composite fibers with unique physical properties for the design of weavable thermoelectric materials and devices. These composite fibers can effectively harness the high σ of carbon materials and the favorable S of semiconductor materials [199]. Through careful design and control of the composition and structure of the composites, it is possible to achieve higher thermoelectric efficiency in their devices, thus more efficiently converting thermal differentials into electrical energy. Carbon materials typically exhibit good mechanical strength and flexibility, enhancing the durability of the composites. This makes them well-suited for applications that require frequent deformation, such as flexible and wearable devices. Additionally, most carbon materials are lighter than traditional inorganic thermoelectric materials, contributing to

reduced device weight, improved comfort, and portability. Furthermore, the production of carbon and semiconductor composite materials can often be achieved using relatively low-cost manufacturing methods such as chemical synthesis and blending processes, thus helping to lower production costs. Moreover, many carbon materials can be obtained through sustainable production methods, reducing dependence on harmful chemicals and scarce resources from a sustainability perspective. Therefore, in recent years, many researchers have turned their attention to this direction for the development of weavable thermoelectric materials and devices.

Considering that weavable thermoelectrics are mostly used at near-room temperatures, the inorganics used for the hybrids are usually Bi_2Te_3 -based materials since they possess promising near-room-temperature ZT s of >1 . Figure 7(a) illustrates the fabrication of CNT yarns with p-type segments deposited by Sb_2Te_3 and n-type segments deposited by Bi_2Te_3 [162]. This is realized by electrochemical deposition of these inorganic chalcogenides on CNT yarns. Figure 7(b) illustrates the conducting mechanism of the as-fabricated yarn [162]. Compared to other deposition methods that require special reactors and high operating temperatures, electrochemical deposition technology can achieve well-controlled structures and high growth rates even with relatively simple equipment and processing conditions, all at room temperature. Additionally, it enables the uniform deposition of thermoelectric materials on a variety of target shapes. Figure 7(c) shows photos of the flexible thermoelectric generator composed of these yarns [162]. This system allows precise control of the temperature gradient using separate modules and water-cooled plates on the hot and cold sides. As the number of p/n pairs increases, device resistance and output voltage are also increased accordingly. Figure 7(d) illustrates wearing the as-fabricated generator for harvesting body heat, and figure 7(e) shows the photo of the wearable thermoelectric generator for power generation [162]. Under indoor conditions at 298 K, a device with 150 p/n pairs connected to the human forearm generates an output voltage of 52.4 mV. This corresponds to a ΔT of 2.9 K between the forearm and the environment, which closely matches the 3.5 K ΔT captured by thermal imaging, as shown in figure 7(f) [162]. Therefore, such a carbon/inorganic fiber-based power generator is promising as a sustainable charger for low-grade wearable electronics.

It should be noted that carbon and inorganic semiconductor composite fibers as weavable thermoelectric materials and devices still face many challenges and controversies. Firstly, selecting the appropriate combination of carbon and inorganic materials to create effective composites is a complex task. Researchers need to carefully choose materials with compatible thermoelectric properties to optimize thermoelectric performance. Achieving optimized thermoelectric performance as much as possible in composite fibers is an ongoing challenge. It is required to balance factors like σ , κ , and S to maximize the ZT . Additionally, establishing a robust and scalable synthesis method is one of the challenges. The production process needs to be cost-effective and suitable for large-scale manufacturing. Also, ensuring stability at high temperatures is crucial for many thermoelectric applications, as some

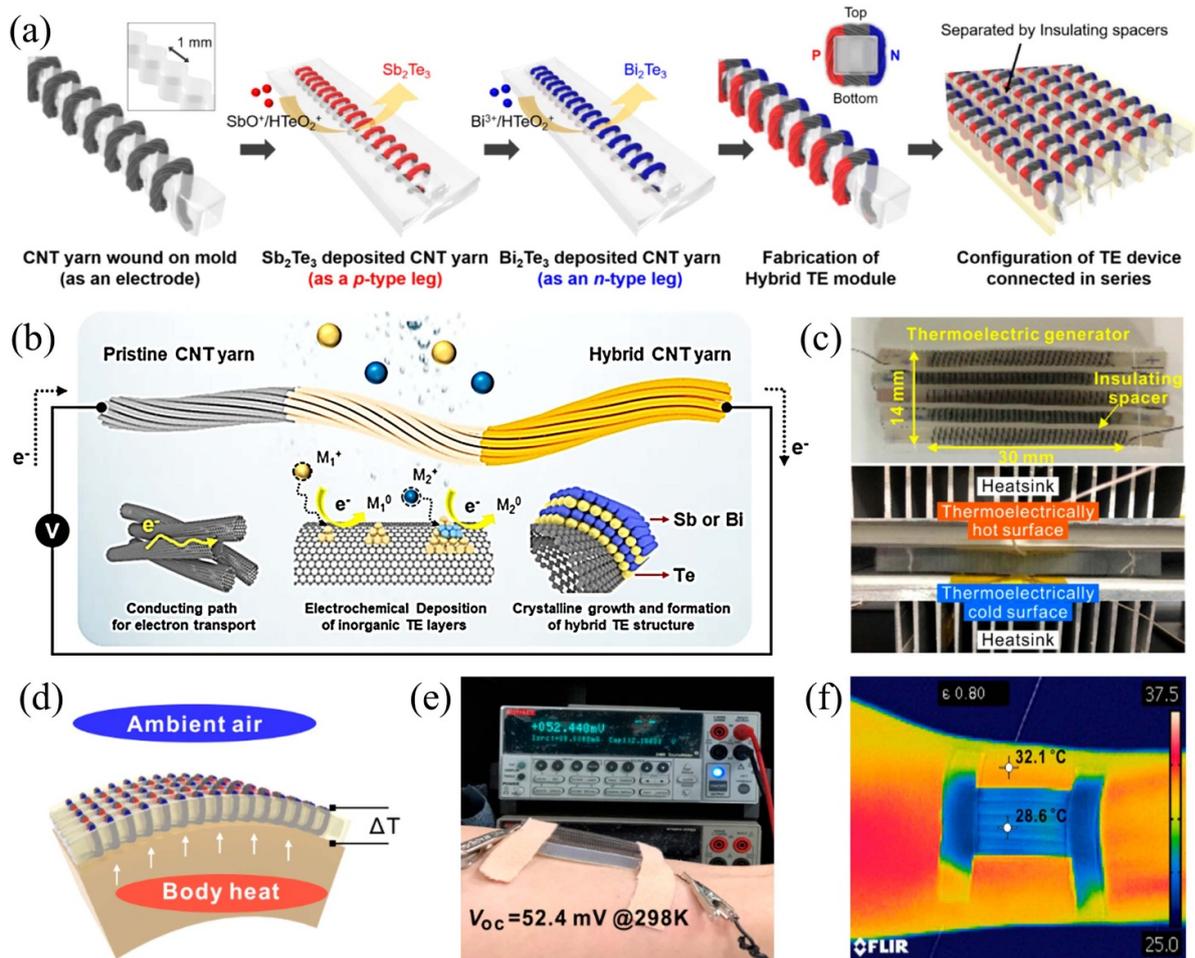


Figure 7. Carbon/inorganic hybrid-based weavable thermoelectrics. (a) Illustration of fabricating CNT yarns with p-type segments deposited by Sb_2Te_3 and n-type segments deposited by Bi_2Te_3 . (b) Illustration of the conducting mechanism of the as-fabricated yarn. (c) Photos of the flexible thermoelectric generator composed of these yarns. (d) Illustration of wearing the as-fabricated generator for harvesting body heat. (e) Photo of the wearable thermoelectric generator for power generation. (f) Corresponding infrared image. Reprinted with permission from [162]. Copyright (2021) American Chemical Society.

carbon materials may degrade or oxidize at elevated temperatures, affecting long-term performance. Some semiconductor materials used in composite preparation may contain toxic elements, raising safety and environmental concerns. Proper disposal and handling are essential. Lastly, producing these composite fibers in an economically efficient manner, especially at scales suitable for practical applications, is a significant issue. High production costs may limit their commercial viability. Overall, carbon and inorganic composite fibers have certain challenges. Therefore, researchers need to continue to work on optimizing these materials to meet the demands in the field of weavable thermoelectrics for smart clothing applications.

8. Carbon/organic hybrid-based weavable thermoelectrics

In addition to conventional inorganic thermoelectric materials, carbon can also hybridize organic conducting polymers to form weavable thermoelectric materials and devices. Carbon and organic conducting polymer composites offer numerous

advantages as weavable thermoelectric materials and devices. Firstly, these composites are typically highly flexible, making them suitable for the fabrication of W-TEDs. They can adapt to various shapes and curved surfaces, which is particularly valuable in wearable electronic devices and textile electronics. Moreover, most carbon and organic polymer composites are relatively lightweight, which is crucial for the comfort and portability of flexible devices [200]. Lightweight materials can reduce the overall weight of the device, making it more suitable for daily use. The performance of these composites can be adjusted and optimized through material design and chemical synthesis to meet the specific requirements of various applications. This flexibility allows for customized fabrication as needed. Additionally, the preparation of carbon and organic polymer composites can often be done using relatively low-cost methods, such as chemical synthesis, which contributes to cost-effectiveness in manufacturing. Furthermore, many carbon materials and organic polymers can be obtained through sustainable production methods, reducing reliance on harmful chemicals and scarce resources. This potential aligns well with sustainability goals. Moreover, most carbon and

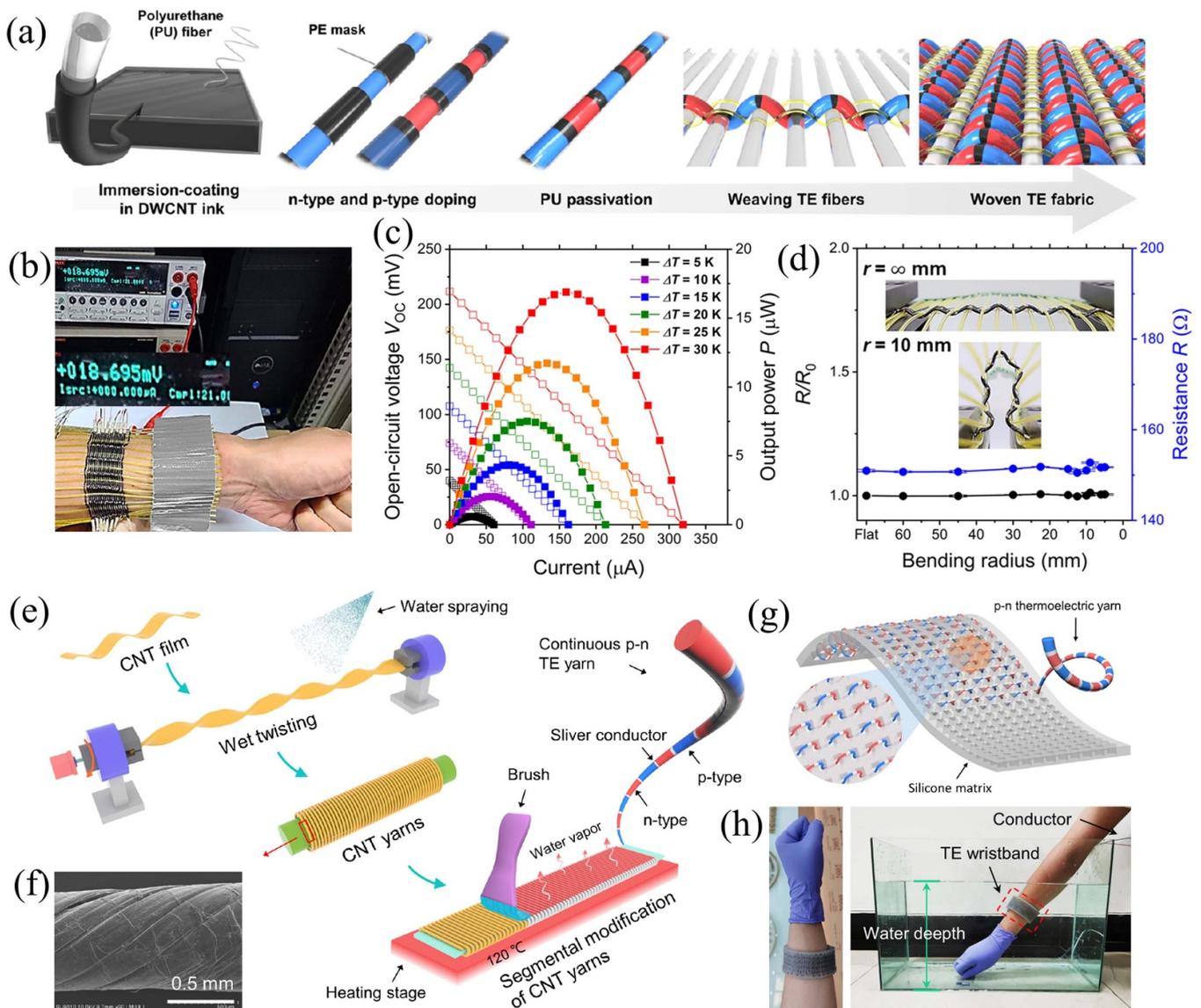


Figure 8. Carbon/organic hybrid-based weavable thermoelectrics. (a) Illustration of fabricating woven thermoelectric fabric composed of hybrid fibers with n- and p-type segments. The hybrid fibers are composed of double-walled CNTs (DWCNTs) coating and polyurethane (PU) core. (b) Photo of wearing the woven thermoelectric fabric for power generation by harvesting body heat. (c) Measured open-circuit voltage V_{OC} and output power P as a function of current under different ΔT s. (d) Normalized resistance R/R_0 and resistance R of the fabric as a function of bending radius. Reprinted from [159], © 2022 Elsevier Ltd. All rights reserved. (e) Illustration of fabricating CNT yarns with n- and p-type segments. (f) Scanning electron microscopy (SEM) image of the as-fabricated CNT yarn. (g) Illustration of the flexible device composed of woven CNT yarns and a silicone matrix. (h) Photo of wearing the device underwater. Reprinted from [157], © 2022 Elsevier B.V. All rights reserved.

organic polymer composites exhibit promising thermoelectric performance within a relatively low-temperature range, which is advantageous for various low-temperature applications. Furthermore, organic polymer materials are typically more biocompatible, making them suitable for applications involving contact with biological systems, such as medical devices or biosensors. Therefore, carbon and organic polymer composites offer unique advantages as thermoelectric fibers.

Till now, there have been many types of carbon/organic hybrid fibers for applying to weavable thermoelectrics, including CNT/PEDOT:PSS yarns [153–155, 160], CNT/PANI fibers [161], PEI-doped CNT yarns [156], WPU

and PVA wrapped SWCNT yarns [158], thermoplastic polyurethane/CNT yarns [152], *etc.* Similar to pure carbon-based and conducting-polymer-based thermoelectric fibers discussed above, carbon/organic hybrid fibers can also be fabricated with p/n segments. Figure 8(a) illustrates the fabrication of woven thermoelectric fabric composed of hybrid fibers with n- and p-type segments [159]. This is a 3D woven fabric with high thermoelectric performance and exceptional mechanical reliability. The hybrid fibers are composed of DWCNTs coating and polyurethane (PU) core. DWCNTs ink coating allows easy deposition of thermoelectric layers onto PU core fibers. The PU passivation layer encapsulated over the coated

fibers enhances the inherent flexibility by redirecting individual DWCNT fibers and improving thermoelectric performance by adjusting heat transfer along the p/n legs. Figure 8(b) shows a photo of wearing the woven thermoelectric fabric for power generation by harvesting body heat, and figure 8(c) shows measured open-circuit voltage V_{oc} and P as a function of current under different ΔT s [159]. The 3D fabric collects heat in-plane and yields a maximum standardized open-circuit voltage of 8.0 mV K^{-1} and a standardized power of $1.1 \times 10^{-4} \mu\text{W K}^{-2}$ per leg. Figure 8(d) shows the measured normalized resistance R/R_0 and resistance R of the fabric as a function of bending radius [159]. The deformability of the weave structure and the elasticity of the fiber induced by passivation collectively ensure reliable fabric mechanical stability, capable of withstanding up to 100% strain from bending and stretching. The thermoelectric woven fabric conforms to the curved human forearm, offering the potential for efficient body heat harvesting, thereby enabling self-powered wearable electronics.

To further enhance the breadth of applications and adaptability to the environment, some research focuses on the packaging of woven TEDs. Figure 8(e) illustrates the fabrication of CNT yarns with n- and p-type segments, and figure 8(f) shows a scanning electron microscopy (SEM) image of the as-fabricated CNT yarn. The fabrication of p-n alternating thermoelectric yarns involves wet-twisting of CNT yarns and segmental modification of the yarns. Importantly, compared to regular twisted yarns without the introduction of deionized water during the twisting process, CNT yarns produced using the wet-twisting method exhibit a denser structure. The capillary action of CNT fiber channels, and the surface tension of wetting water droplets, collectively contribute to the good performance during twisting. Figure 8(g) illustrates the flexible fabric device composed of woven CNT yarns and a silicone matrix [157]. The design of this fabric is based on a compliant packaging solution for thermoelectric generators, ensuring the proper alignment of thermal gradients based on the infrared reflection effect. This design increases thermal insulation at the hot side due to enhanced infrared radiation reflection and reduces radiative heating at the cold side. When exposed to the same heat source, it results in a significant increase in the temperature gradients, leading to an approximately 35% increase in the output voltage compared to that of the unencapsulated version. Figure 8(h) shows a photo of the device underwater [157]. Thanks to its waterproof enclosure, this thermoelectric wristband proves to be reliable in various real-life activities such as running and swimming [157].

Currently, carbon and organic polymer composite fibers still face several challenges and controversies in both research and applications. Firstly, despite their numerous advantages such as flexibility and lightweight nature, these composites often exhibit relatively low conductivity and thermoelectric performance. Therefore, continuous efforts are needed to enhance their thermoelectric properties while retaining their other advantages to improve the energy conversion efficiency of the woven device. Also, the quality of interfaces within carbon and organic polymer composites is crucial for the transfer of electrons and phonons. Achieving high-quality interfaces is

a complex task that requires overcoming potential defects or barriers at the interface to maximize performance. In addition, organic materials may undergo degradation under high temperatures and extended use, potentially reducing their stability and lifespan. Therefore, methods to enhance the stability of these materials need to be explored to meet the requirements of practical applications. Moreover, the production of high-performance carbon and organic polymer composites typically involves complex synthesis and processing steps, which can increase costs. Therefore, there is a need to develop more cost-effective and efficient manufacturing methods to enable commercial production. Some organic polymer materials may also be sensitive to environmental factors such as humidity, oxygen, and ultraviolet light, which can lead to a decline in performance or failure. Additionally, some components used in the preparation of these composites may contain toxic elements, raising concerns about safety and environmental impact. Therefore, proper disposal and handling are important considerations. Finally, while these materials show promise in laboratory settings, successfully commercializing them and scaling up production remain challenging tasks. High production costs and market competition are obstacles that need to be overcome. Overall, addressing these challenges and controversies is essential for realizing the potential of carbon and organic polymer composite fibers as viable options in the field of weavable thermoelectrics.

9. Organic/inorganic hybrid-based weavable thermoelectrics

Organic/inorganic composite materials are a key focus in the field of thermoelectric research [201]. Inorganic nanomaterials and organic polymer composites offer several advantages, which are beneficial for acting as weavable thermoelectric materials and devices. Inorganic nanomaterials, such as Bi_2Te_3 nanoparticles or Te NWs [166], typically exhibit excellent σ and S , as well as low κ . This makes them highly effective as inorganic fillers for tuning the overall thermoelectric performance of hybrid materials. Furthermore, the performance of these materials can be customized by adjusting the size, shape, and combination of nanostructures. Organic conducting polymer materials, when used as matrices, tend to be flexible, ensuring the overall flexibility of composites, and they often have low κ . Additionally, at the interfaces between organic and inorganic materials, there is typically an energy-filtering effect that filters low-energy charge carriers, enhancing the S while maintaining high σ [201]. Therefore, these composite fibers hold great promise in the field of weavable thermoelectric materials and devices due to their ability to combine the strengths of both organic and inorganic materials, offering opportunities for customized performance tuning.

Till now, there have been many types of inorganic/organic hybrid fibers for applying to weavable thermoelectrics, including polyvinylpyrrolidone (PVP)-assisted Bi_2Te_3 fibers [165], PEDOT:PSS/Te-NWs [163, 164], PEDOT:PSS/polyvinyl alcohol (PVA)/Te ternary composite fibers [166], *etc.* The

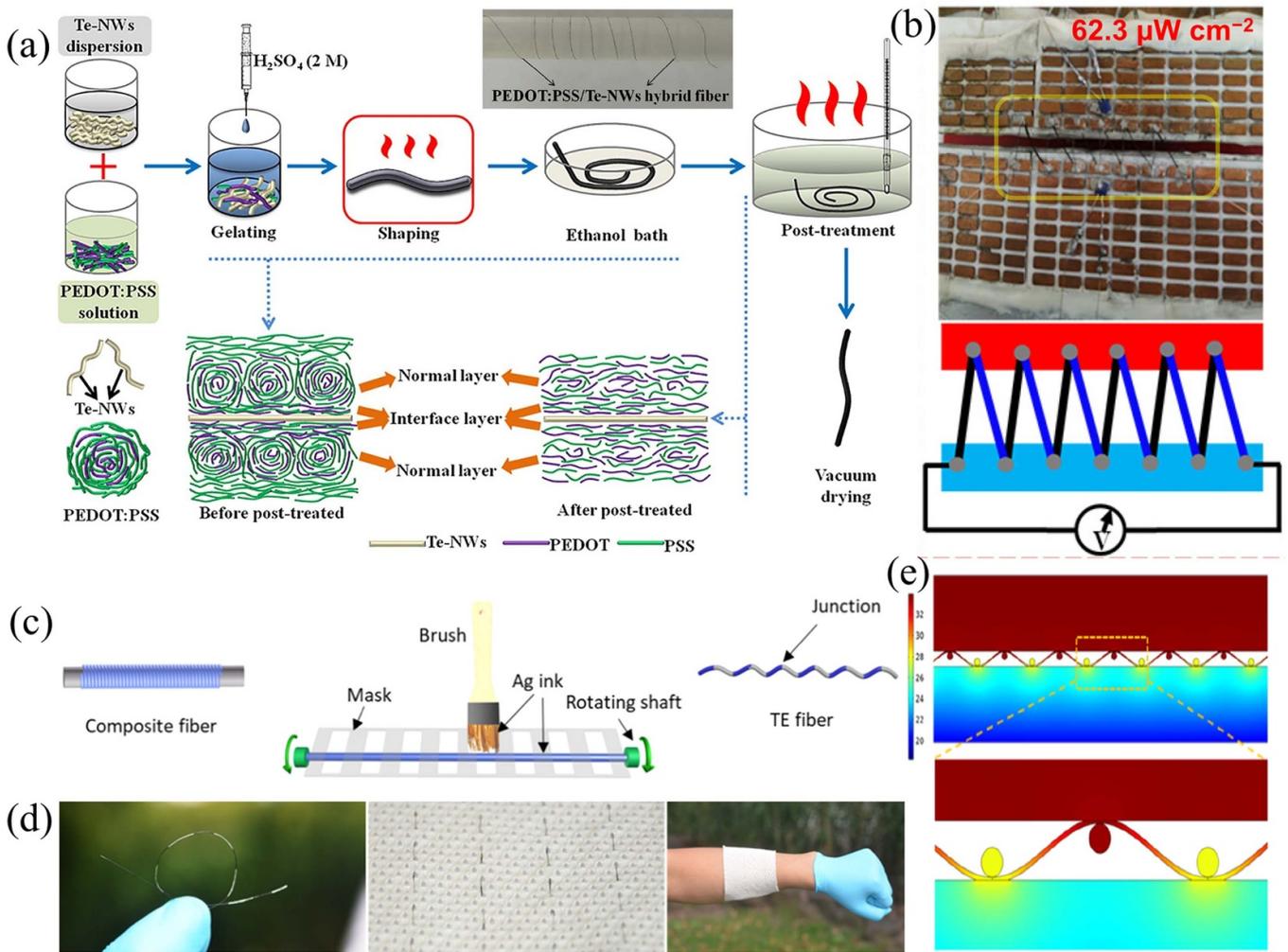


Figure 9. Organic/inorganic hybrid-based weavable thermoelectrics. (a) Illustration of fabricating hybrid fibers composed of PEDOT:PSS and Te NWs. (b) Photo of the woven device composed of hybrid fibers. Reprinted from [163], © 2020 Elsevier B.V. All rights reserved. (c) Illustration of fabricating composite fibers with n- and p-type segments. (d) Photos of the composite fibers that can be woven into a fabric for wearable applications. (e) Illustration of the thermal distribution. Reprinted with permission from [164]. Copyright (2020) American Chemical Society.

solution-based method is one of the most common ways to fabricate organic/inorganic hybrid fibers. Figure 9(a) illustrates the fabrication of hybrid fibers composed of PEDOT:PSS and Te NWs [163]. Utilizing solvent post-treatment is an effective strategy to enhance the thermoelectric performance and mechanical properties of manufactured hybrid fibers. After treatment with H_2SO_4 , the PEDOT:PSS/Te-NWs hybrid fibers containing 50 wt% Te-NWs achieved a high $S^2\sigma$ of up to $17.8 \mu\text{W m}^{-1} \text{K}^{-2}$ and exhibited good mechanical stability. Figure 9(b) shows a photo of the woven device composed of hybrid fibers [163]. This device features six pairs of p-n fiber legs and exhibits a power density of $\sim 60 \mu\text{W cm}^{-2}$ at a ΔT of 40 K. Another case is highly integrable thermoelectric fiber. Figure 9(c) illustrates the fabrication of composite fibers with n- and p-type segments [164]. The fiber was fabricated by a wet-spinning process, and the p-type segments are composed of PEDOT:PSS/Te-NWs hybrids. This thermoelectric fiber demonstrates ideal thermoelectric performance including a high $S^2\sigma$ of $78.1 \mu\text{W m}^{-1} \text{K}^{-2}$. Figure 9(d) shows

the photos of the composite fibers that can be woven into a fabric for wearable applications [164]. This thermoelectric fabric demonstrates a high specific power of $9.48 \mu\text{W g}^{-1}$, excellent mechanical flexibility, and outstanding integrability. Figure 9(e) illustrates the thermal distribution within the fabric [164]. The use of finite element analysis and simulations helps explain the direction of heat transfer and the way the temperature difference between human skin and the surrounding environment is formed. This plays a role in understanding the device performance.

Similarly, inorganic nanomaterials and organic polymer composites, when used as weavable thermoelectrics, still face several issues in current research and applications. The primary challenge lies in ensuring that the interface between organic polymers and inorganic nanomaterials in composites maintains high quality, which is essential for promoting the transfer of electrons and phonons, and this remains a complex task. The fundamentals for the energy filtering effect are still not clear. Defects or barriers at the interface

can significantly impact material performance. Therefore, further in-depth research and engineering efforts are required to enhance interface compatibility. Also, organic polymer materials are typically prone to degradation at high temperatures or during prolonged usage, potentially reducing their stability and lifespan. The preparation of high-performance inorganic nanomaterials and organic polymer composites often involves complex synthesis and processing steps, which can increase costs. Some organic polymer materials are more sensitive to environmental factors such as humidity, oxygen, and ultraviolet radiation than inorganic materials, which may lead to performance degradation or failure. Additionally, some components used in the preparation of these composite materials may contain toxic elements, which should be avoided. Finally, while these materials have demonstrated potential in laboratory settings, successfully commercializing and scaling up production still presents significant challenges.

10. Fabrics as thermoelectric materials for weavable thermoelectrics

In the previous discussions, the research focus has primarily been on 1D weavable thermoelectric fibers, with relatively fewer studies on fabrics used as flexible substrates. Fabrics used as flexible substrates can also be functionalized, especially for thermoelectric purposes, to further enhance the overall performance of W-TEDs. For example, common design approaches include modifying traditional wool or synthetic fabrics and depositing thermoelectric layers onto them using solution-based or deposition methods [176–186]. Additionally, some research explores the direct use of thermoelectric fabrics as 3D thermoelectric legs, functioning similarly to 1D weavable thermoelectric fibers.

The fabrics can serve as substrate or template materials for depositing thermoelectric materials as coatings. For the case of direct use of thermoelectric fabrics as 3D thermoelectric legs, figure 10(a) illustrates filled, 10% filled bulk devices, and a network-based flexible device, respectively [181]. Compared with conventional devices with bulk materials, the ones with fabric networks can exhibit both excellent elasticity and good thermoelectric performance. To achieve this goal, figure 10(b) illustrates the composition of the Ag_2Se network, which is *in-situ* synthesized by silvering and then selenizing [181]. Figure 10(c) is the photo of a large-scale Ag_2Se network. Such a network shows a light weight of 0.28 g cm^{-3} , a low κ of $0.04 \text{ W m}^{-1} \text{ K}^{-1}$, a moderate softness of 0.03 MPa , and a high elongation of $>100\%$ [181]. The as-fabricated network can be made into different shapes to benefit the designs of different devices, as indicated by figure 10(d) [181]. Figure 10(e) shows some photos of a wearable thermoelectric generator composed of an Ag_2Se network, indicating good wearability [181]. Figure 10(f) also shows the measured and calculated power density of the generator as a function of current [181]. A high output power of $4 \mu\text{W cm}^{-2}$ can be achieved in such a device, which is a promising value for applying to wearable electronics. Other works reported 3D-printed $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ open-cell honeycomb structure features a high surface area,

enabling high heat transfer rates throughout the heat pipe, with negligible impact on the liquid flow [184], therefore acting as promising porous thermoelectrics for *in-situ* energy harvesting.

In addition to the above thermoelectric fabric, there have been reports on the development of coated thermoelectric fabrics, attempting to coat fabrics with PEDOT:tosylate and CuI nanocrystals [177]. Various types of fabrics such as cotton, nylon, NYCO, polyester, nomex, modified acrylic, *etc.*, have been used as substrates for manufacturing different composite materials. The conclusion drawn was that NYCO composed of 50% cotton and 50% nylon provided the highest thermoelectric performance. CuI nanocrystals enhanced the thermoelectric performance of the composite material, and the device power density reached 3.68 nW cm^{-2} at a ΔT of 12 K. When worn by a person, the device was capable of generating a P of 12.4 nW . Also, a thermoelectric fabric coated by CNT/PVP hybrids was prepared using a simple ultrasonic coating method and further extended for its application in self-powered temperature/strain sensing [176]. PVP was used to disperse CNTs, enhancing the adhesion between the fabric and CNTs, thereby maintaining stable thermoelectric performance under repeated bending and stretching. The proposed self-powered temperature sensor showed significant potential in temperature recognition and respiratory monitoring. Similarly, SnS and SnSSe_x ($x = 0.05, 0.075, \text{ and } 0.1$) samples were synthesized through a hydrothermal method and coated onto carbon fabric using a drop-casting technique for flexible thermoelectric applications [185]. A $S^2\sigma$ of $1.56 \mu\text{W m}^{-1} \text{ K}^{-2}$ was achieved for $\text{SnSSe}_{0.1}$ at 373 K. Other works also reported thermoelectric fabrics such as electrochemical polymerization of PEDOT on felt fabrics [180], worm-Like PEDOT: Tos-coated polypropylene (PP) fabrics [182], ordered Bi_2Te_3 crystals anchored on carbon fiber networks [186], electrospraying CNTs on poly (lactic acid) (PLA) fabric [179], solvent-assisted synthesis of Ag_2Se and Ag_2S nanoparticles on carbon fabric [178], *etc.*

Some works also focused on weaving thermoelectric fibers into 2D films for thermoelectric applications. For example, CNT/PVP/PU composite films were fabricated by electrospinning technology and air pressure spraying [183]. This material combines high breathability and stretchability, close to that of pure PU nanofiber fabric ($\sim 250\%$). Additionally, PVP simultaneously enhances the dispersion of CNTs and acts as an interface binder between CNTs and the elastic PU scaffold. Consequently, even after 1000 bending cycles, the σ and S remain unchanged. Moreover, self-powered sensors were successfully fabricated for finger temperature and speech conversion, as well as joint motion detection to optimize athlete performance.

11. Fabrics as substrates for weavable thermoelectrics

As discussed above, fabric is an essential part of human life, and by combining thermoelectric principles with textile technology, thermoelectric fabrics that are soft, three-dimensionally conformable, breathable, moisture-wicking,

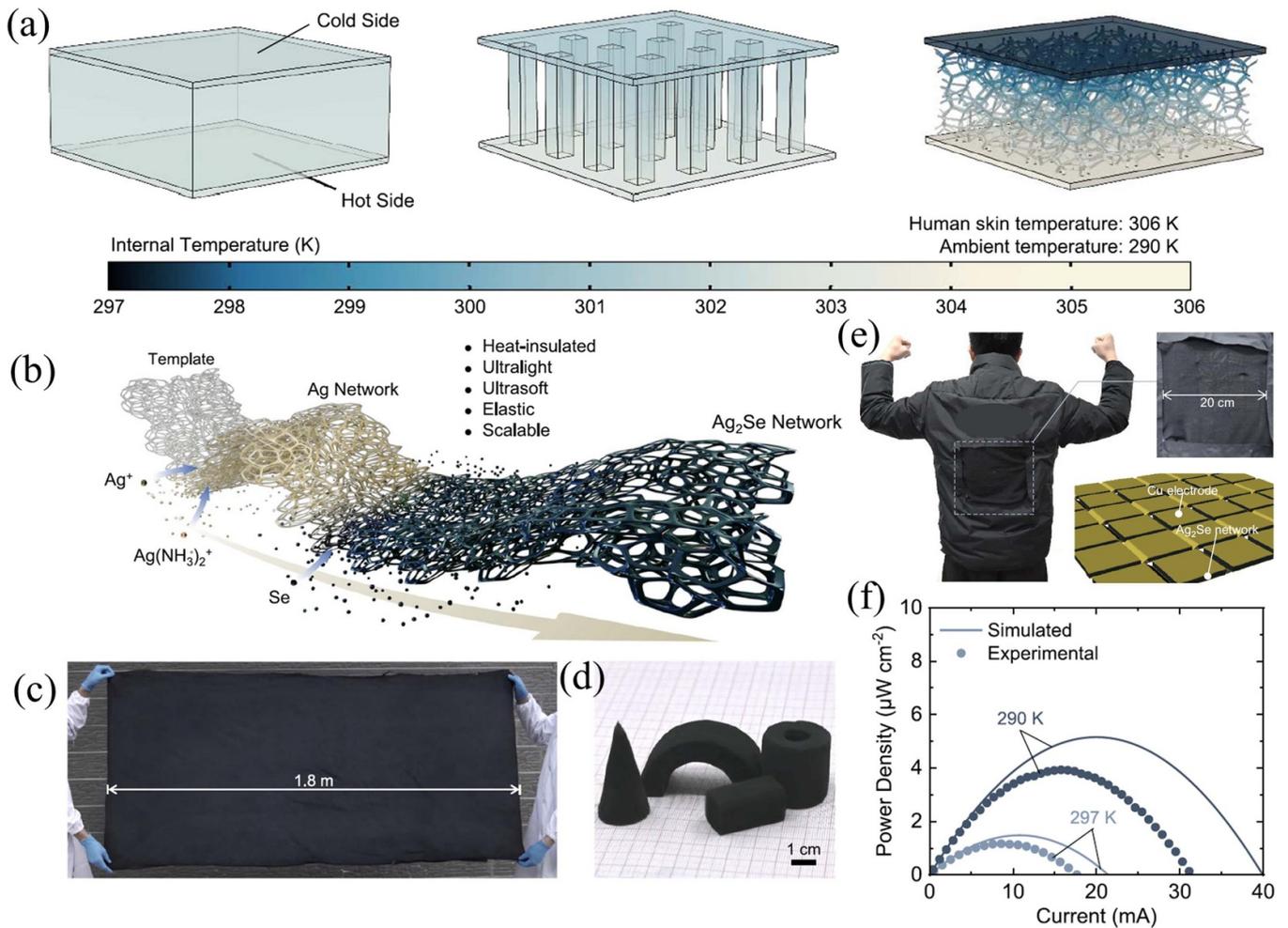


Figure 10. Fabrics as thermoelectric materials for weavable thermoelectrics. (a) Illustration of filled, 10% filled bulk devices, and a network-based flexible device. (b) Illustration of the composition of the Ag_2Se network. (c) Photo of a large-scale Ag_2Se network. (d) Photo of the Ag_2Se network made into different shapes. (e) Photos of wearable thermoelectric generator composed of Ag_2Se network. (f) Measured and calculated power density of the generator as a function of current. Reproduced from [181]. CC BY 4.0.

capable of temperature regulation, and energy-harvesting have been created. These thermoelectric fabrics hold significant application value in the regulation of body surface temperature and the collection of thermal energy. In addition to using conventional fabric as a substrate or support material for the deposition of thermoelectric materials, some fabrics are used directly as support materials for TEDs or other matching functional devices. To achieve specific goals, these fabrics require careful selection based on factors such as shape, porosity, and material, and may also need further modifications. Figure 11(a) illustrates the structure of a wearable device with embedded bulk thermoelectric materials, and figure 11(b) shows the body surface temperature changes as a function of cooling time by wearing the device [174]. This is a scalable manufacturing method for thermoelectric fabrics, achieved by directly weaving inorganic bulk materials into the fabric using a semi-automatic weaving machine. This method allows for continuous weaving of large areas (1550 cm^2), washable, and mechanically durable thermoelectric fabrics. Through systematic optimization of its thermal resistance, the thermoelectric

fabric can achieve a cooling temperature of 11.8 K on the human body surface at an ambient temperature of 34°C , with a cooling power of 553.7 W m^{-2} . It can also generate a thermoelectric power density of 6.13 W m^{-2} at a ΔT of 25 K. In a scenario where it is worn by a person, this thermoelectric fabric can stably power small mobile phone lithium batteries under a self-established ΔT of 15 K. Other works also reported patterned elastic fabric substrate with embedded rigid thermoelectric cuboids, together with serpentine structured cloth electrodes to form whole fabric-assisted TEDs for wearable electronics [173]. Such a device showed a large peak P of $64.1 \mu\text{W}$ at a ΔT of 33.24 K.

Sometimes, the fabric should be modified to benefit the designs of complex energy harvesting systems. Figure 11(c) illustrates the structure of a fabric-based energy harvester composed of thermoelectric generators and triboelectric nanogenerators [175]. This composite device is manufactured through a three-layer sequential stacking. At the bottom, an aluminum-coated fabric can come into direct contact with human skin. In the middle, the original fabric adheres to

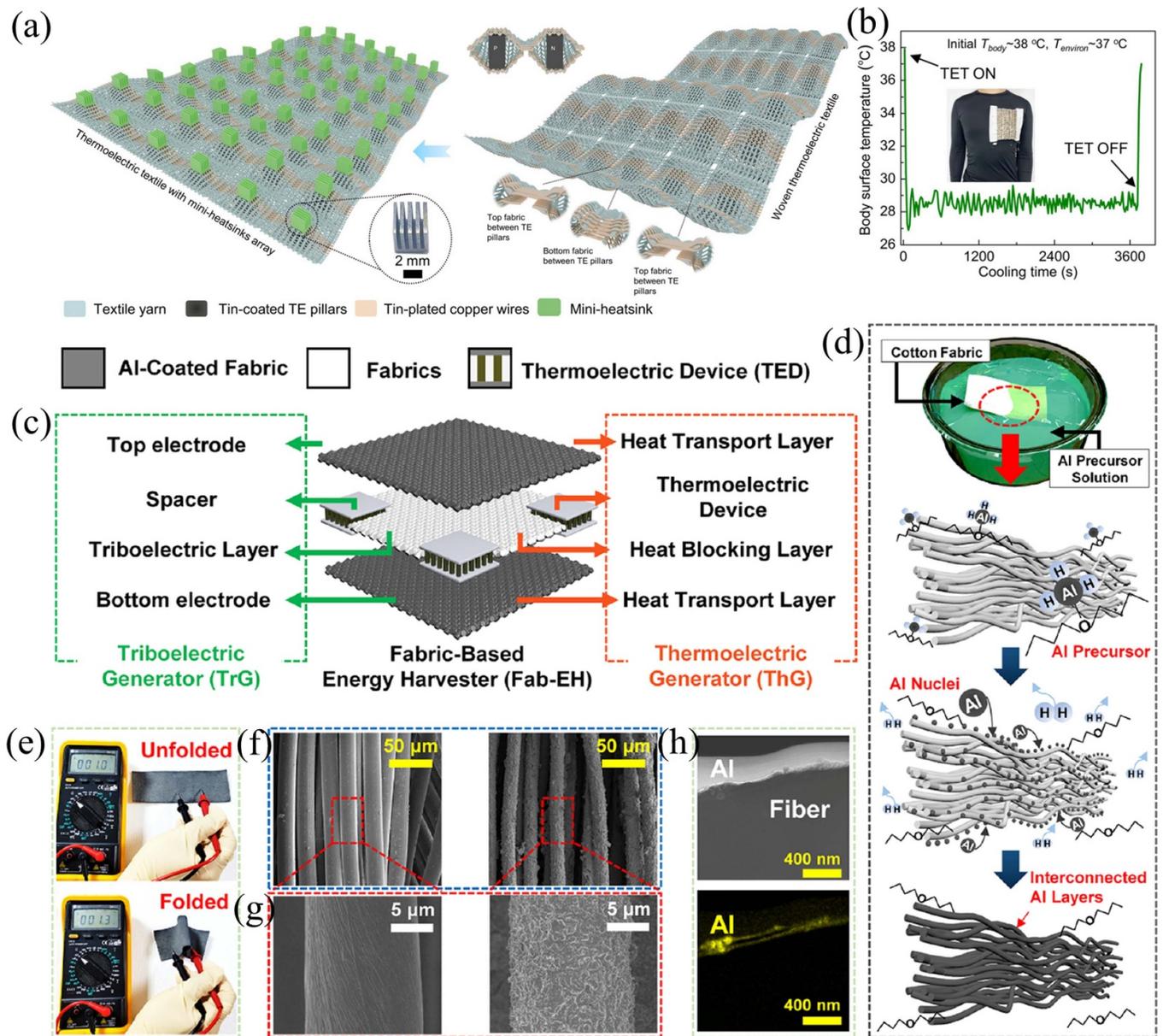


Figure 11. Fabrics as substrates for weavable thermoelectrics. (a) Illustration of the structure of a wearable device with embedded bulk thermoelectric materials. (b) The body surface temperature changes as a function of cooling time by wearing the device. Reproduced from [174] with permission from the Royal Society of Chemistry. (c) Illustration of the structure of fabric-based energy harvester. (d) Illustration of coating Al on cotton fabric. (e) Photos of unfolded and folded Al-coated fabric. (f) Low- and (g) high-magnification SEM images of the fabric before and after Al-coating. (h) Transmission electron microscopy (TEM) image (top) and energy-dispersive x-ray spectroscopy (EDS) map (bottom) to confirm the existence of Al coating. Reprinted from [175], © 2022 Published by Elsevier Ltd.

the commercial TED, and at the top, there is another piece of aluminum-coated fabric exposed to the air. Each fabric layer and the four TEDs serve two functions. To functionalize the composite device, the fabric should be coated with high-quality aluminum. Figure 11(d) shows the aluminum plating technique used to prepare aluminum-coated fabric [175]. The fabric is made by weaving threads, which are finer bundles of fibers. Aluminum plating technology uniformly forms an aluminum film on the surfaces of the fabric on all sides. Due to the uniformity and conformity of the aluminum film, high σ and κ are achieved while maintaining the texture of the original fabric. Optical photographs of the conductive aluminum-coated

fabric, as well as SEM images of several types of cotton fibers before and after the aluminum plating process, are shown in figures 11(e)–(g) [175]. Even when the conductive aluminum-coated fabric is folded, its σ remains largely unchanged. All fibers constituting the fabric received a good coating due to the coating technology being based on a solution process. To determine the thickness of the coated aluminum, transmission electron microscopy (TEM) images and energy-dispersive x-ray spectroscopy (EDS) mapping images were obtained, as shown in figure 11(h) [175]. The measured thickness of the coated Al is approximately $0.2 \mu\text{m}$, which is only 1.3% of the diameter of the fiber. Therefore, by using aluminum-coated

fabric material as the electrodes for triboelectric nanogenerators and the heat transport layer for thermoelectric generators, the manufactured composite device effectively harvests electrical energy from body motion and body heat. The composite device charges a storage capacitor with a capacitance of 3.3 mF to 3 V within 240 s using a designed voltage-boosting system. Additionally, the collected energy can partially charge a smartphone.

In conclusion, temperature regulation through thermoelectric fabrics has significant implications both for human health and for reducing energy consumption. Achieving a two-degree temperature regulation for the human body can save approximately 20% of energy consumption in systems such as air conditioning and ventilation [174]. Thermoelectric fabrics offer advantages such as portability, noiselessness, absence of moving parts, and no additional energy input requirements. They harness the temperature gradients between the human body and the environment to power various portable electronic devices, making them valuable in the fields of smart wearables and energy storage. While the cost of inorganic thermoelectric materials used in these fabrics can be relatively high, thermoelectric fabrics can be environmentally recycled by physically disassembling them, allowing for repeated use and cost reduction, thus maximizing economic benefits.

12. NWs for weavable thermoelectrics

NWs typically have only nanoscale dimensions [202], making them challenging to weave into thermoelectric fabrics. However, understanding their growth mechanisms and harnessing their properties can guide future applications in woven TEDs. Additionally, many NWs can be synthesized as fillers for composite thermoelectric fibers, ensuring ongoing research in NWs applications.

Figures 12(a)–(c) shows the fabrication of porous Si NWs, including patterning metals by the nanoimprint lithography method, fabricating NWs by chemical etching assisted by metal, and a post-doping process [167]. The as-synthesized large-area, wafer-level porous silicon NW arrays possess ultrathin silicon grain sizes of approximately 4 nm. Figure 12(d) is an SEM image of the as-fabricated Si NW [167]. A ZT of 0.31 at room temperature and a high ZT of 0.71 at 700 K can be achieved. By designing an advanced suspended microdevice platform, the measurement accuracy can be improved, and the measurement temperature range can be further extended. In terms of the case of synthesizing NWs as fillers for composite thermoelectric fibers, figure 12(e) illustrates the fabrication of Ag_2Se NWs coated by polyaniline (PANI) based on aqueous solutions [168]. For the composites with 65 wt% NW, a high $S^2\sigma$ of approximately $196.6 \mu\text{W m}^{-1} \text{K}^{-2}$ was achieved at 300 K. After 1000 bending cycles, the initial $S^2\sigma$ was only decreased by about 8%, demonstrating high flexibility. A six-leg TED was assembled using composites, which generated approximately 15.4 mV of voltage and 835.8 nW of maximum P at a ΔT of around 30 K. Additionally, the flexible device exhibited a high power density, approximately 2.33 W m^{-2} at a ΔT of 30 K.

NWs as weavable thermoelectric materials come with various potential issues and challenges in the current stage. These include difficulties in controlling their sizes, high fabrication costs, limited stability, the complexity of selecting suitable NW materials to achieve desired thermoelectric performance, low scalability, significant environmental impact, and poor material durability. Therefore, in the development and application of NWs as weavable thermoelectric materials, addressing these issues and challenges is essential to maximize their potential applications [203].

13. Weavable thermoelectric thin films

In recent years, the majority of reported F-TEDs have been based on 2D flexible films [67]. These devices are often in a horizontal 2D configuration, where thin films are simply assembled into clothing or flexible substrates, as shown in figures 13(a) and (b), from which an F-TED composed of n- and p-type SWCNT films and Kapton substrate is shown for integrating with fabric [171]. As a result, they typically maintain a relatively small temperature difference during wear. This is one of the reasons for researching and developing 3D weavable thermoelectric fiber materials and devices. However, it is worth noting that through specific device designs, 2D films can also be used to assemble 3D W-TEDs. This allows for the possibility of maintaining higher temperature differentials while matching them with flexible heat sinks to further stabilize the temperature difference for consistent power output.

To utilize 2D flexible films for a 3D device configuration, one typically needs to consider designing unique device structures. Figure 13(c) illustrates the fabrication of fiber-shaped thermoelectric generators [170]. This is a flexible rolled thermoelectric module with a radial heterojunction interlayer, featuring a high-density p-n films achieved through the rolling patterning of CNT films (active material layer) and cellulose nanofiber membrane (insulation layer). When the length of the rolled module is 2 cm, it achieves high integration with approximately 3.32 mm^3 per thermoelectric pair, and a high output voltage density of $\sim 65.4 \text{ mV cm}^{-2}$ at a ΔT of 41 K. Furthermore, fiber-shaped TEDs were designed by interconnecting rolled modules, featuring axial and radial p-n junctions, achieving an open-circuit voltage density of $0.176 \text{ mV cm}^{-1} \text{K}^{-1}$. Additionally, fiber-shaped TEDs can be woven into thermoelectric fabrics using commercial yarns to harvest body heat through alternating rolling modules. Other works reported the design of flexible spring-shaped TEDs, as illustrated by figures 13(d) and (e) [169]. This is a typical 3D spring-like TED with a dual-layer elastic structure and gaps. The novel device architecture inherits good flexibility and compressibility, and it also possesses the capability to harvest waste heat under vertical temperature gradients. Detailed analysis of the thermal model shows three optimizations compared to traditional TEDs, namely enhancing contact for more efficient interfacial heat transfer, constructing insulating gaps to impede heat transfer across the entire device body, and efficient heat dissipation with a heat sink to facilitate rapid heat transfer to the environment. The

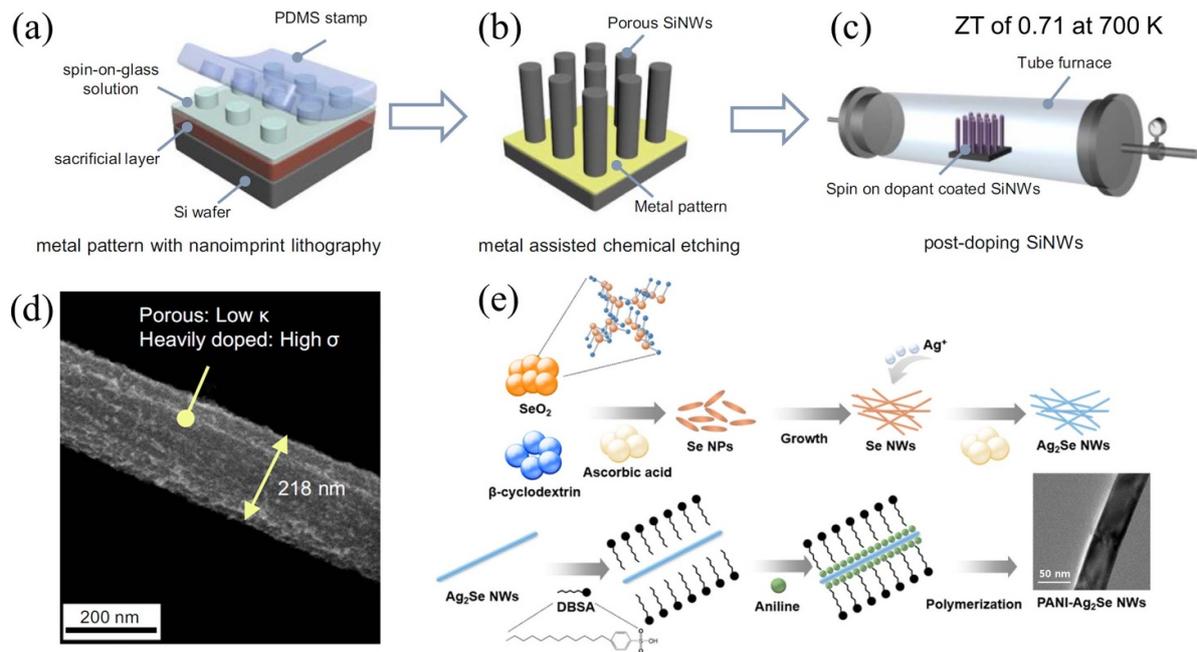


Figure 12. Nanowires for weavable thermoelectrics. (a) Illustration of patterning metals by the nanoimprint lithography method. (b) Illustration of fabricating Si NWs by chemical etching assisted by metal. (c) Illustration of post-doping Si NWs. (d) SEM image of the as-fabricated Si NW. Reproduced from [167]. CC BY 4.0. (e) Illustration of fabricating Ag_2Se NWs coated by polyaniline (PANI). Reprinted from [168], © 2021 Elsevier B.V. All rights reserved.

manufactured TED, under a vertical gradient of 30 K, provides a high output P of 749.19 nW with only three pairs of p-n thin films, equivalent to a high output power density of $416.22 \text{ nW cm}^{-2}$, which is comparable to other reported F-TEDs [67]. Similarly, figure 13(f) illustrates the structure of a wearable thermoelectric generator composed of PEDOT:PSS thin films and Al wires [172]. When applied to the wrist, it exhibits a V_{oc} of 1.46 mV. Subsequently, the device was tested at higher temperatures, and characterization results show that the device achieves a maximum V_{oc} of 5.15 mV at a ΔT of 80 °C. In this situation, the device provides a maximum output P of approximately 2.4 nW and an output power density of $\sim 1.5 \text{ nW cm}^{-2}$.

It is worth noting that whether using 1D thermoelectric fibers, 2D thermoelectric films, or 3D thermoelectric fabrics, there are still several issues and challenges associated with 3D W-TEDs. Firstly, designing and fabricating W-TEDs with complex 3D structures require advanced processing techniques and equipment, which increases the complexity and cost of fabrication. Secondly, the intricate 3D structures often involve complex heat transfer paths and current distributions, which can impact the device performance and efficiency. Detailed design and optimization work are necessary to achieve efficient energy conversion. Additionally, integrating 3D W-TEDs into practical applications may require addressing complex integration issues among electronic components, sensors, and power sources. Lastly, in the fabrication and handling of 3D weavable devices, environmental factors, and material sustainability need to be considered to reduce adverse environmental impacts. Addressing these issues and challenges may require interdisciplinary research

and engineering efforts to advance the development and application of 3D W-TEDs.

14. Weavable integrated systems

To further expand the application areas of weavable thermoelectric materials and devices, as well as to achieve multi-functional integration goals such as electronic skin and smart textiles for human thermal management, weavable thermoelectric materials and devices often need to be used in conjunction with other wearable functional devices. This involves interdisciplinary challenges and can be quite complex. The types of wearable functional devices vary widely and can range from simple devices like current/voltage regulators (e.g. amplifier) [103], micro-supercapacitors [150], triboelectric nanogenerators [175, 204], photovoltaic generators [205], moisture generators [206], ferroelectric generators [204, 207], to more complex objectives such as creating a human sensing network [208], luminous fabrics [209], smart eye masks [210], intelligent gloves [211], sleep monitoring/smart beddings [212, 213], electronic clothing [214], and more.

In general, there are not many cases where weavable thermoelectric materials and devices are used in integration with other wearable functional devices. Figure 14(a) illustrates the assembling procedures of the piezo/thermoelectric bimodal tactile sensor array, and figure 14(b) illustrates the structure of the integrated device [189]. Here, a 3D processing technique was employed, combining laser manufacturing and screen printing, to build a vertical architecture pressure/temperature dual-peak active sensor using pure organic

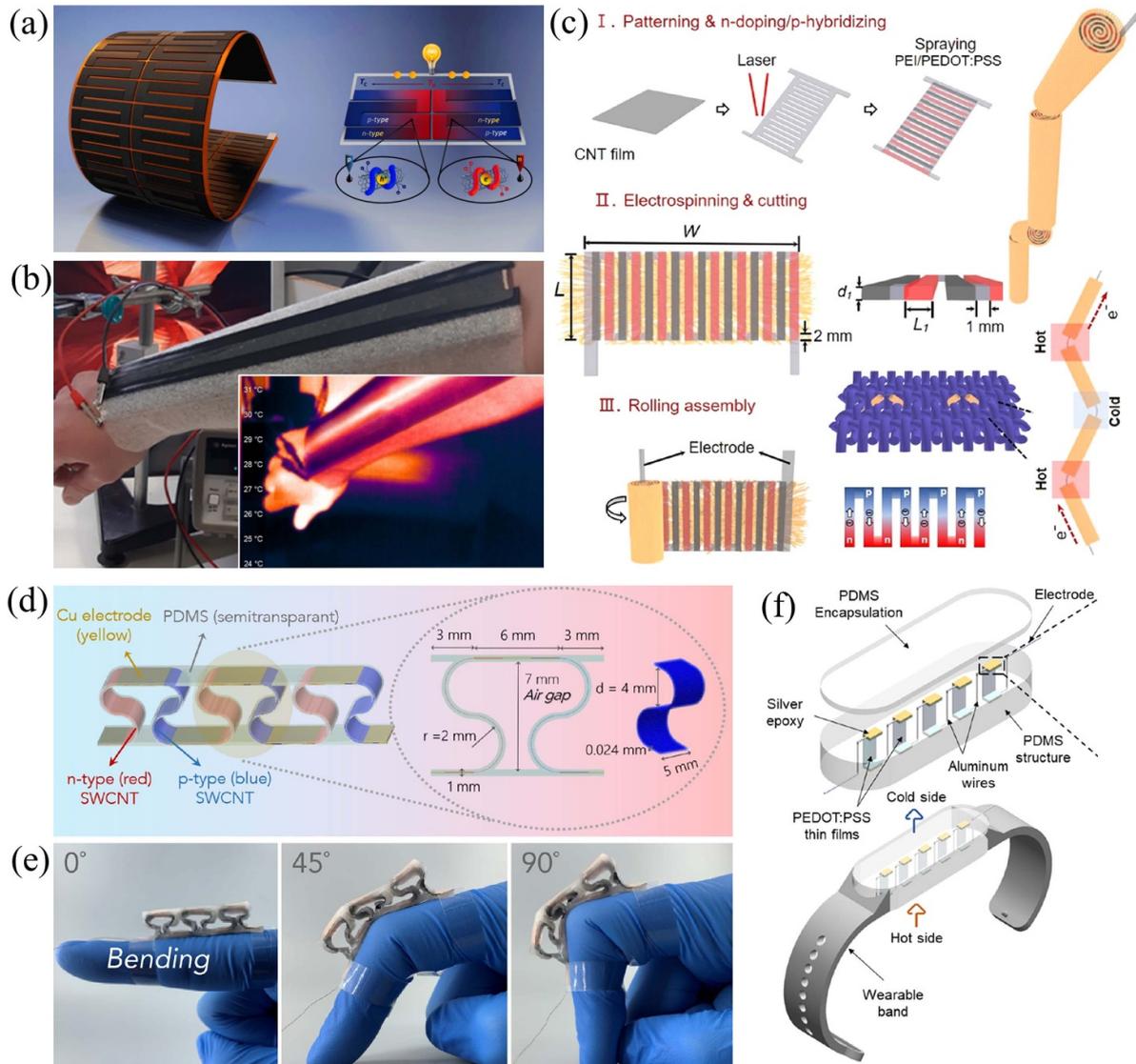


Figure 13. Weavable thermoelectric thin films. (a) Illustration of a flexible TED (F-TED) composed of n- and p-type SWCNT films and Kapton substrate for integrating with fabric. (b) Photo and corresponding infrared image of wearing the fabric with an embedded device. Reprinted with permission from [171]. Copyright (2021) American Chemical Society. (c) Illustration of fabricating fiber-shaped thermoelectric generators. Reprinted from [170], © 2022 Elsevier Ltd. All rights reserved. (d) Illustration of the structure of the flexible spring-shaped TED. (e) Photos showing the flexibility of the device. Reprinted from [169], © 2021 Elsevier Ltd. All rights reserved. (f) Illustration of the structure of a wearable thermoelectric generator composed of PEDOT:PSS thin films and Al wires. Reprinted from [172], © 2022 Published by Elsevier Ltd.

functional materials, namely, piezoelectric poly(vinylidene fluoride-trifluoroethylene) and thermoelectric polyaniline-based composites. Figure 14(c) shows photos that indicate its foldability and wearability, and figure 14(d) shows the change in voltage during the bending of the device [189]. This sensor converts pressure and temperature stimuli into two independent electrical signals without interference, exhibiting high-temperature sensitivity of $109.4 \mu\text{V K}^{-1}$ and a fast response time of only 0.37 s. It also demonstrates excellent pressure sensitivity over a wide range from 100 Pa to 20 kPa. Figure 14(e) compares the sensing results when touching 0°C (left) and 50°C (right) water by wearing the integrated device [189]. Hence, this research represents a step towards multifunctional flexible electronic devices for electronic skin

applications. Considering electronic skin as a human-machine interface with vast prospects in future personal health monitoring and conferring tactile capabilities to robots, such interdisciplinary frontier research is crucial.

15. Future perspectives

At the current stage, weavable thermoelectric materials and devices still face key challenges in practical applications, including (1) effectively capturing the direction of heat flow from human environmental temperature gradients to increase power density; (2) enhancing the thermoelectric performance and stability of thermoelectric materials, especially n-type

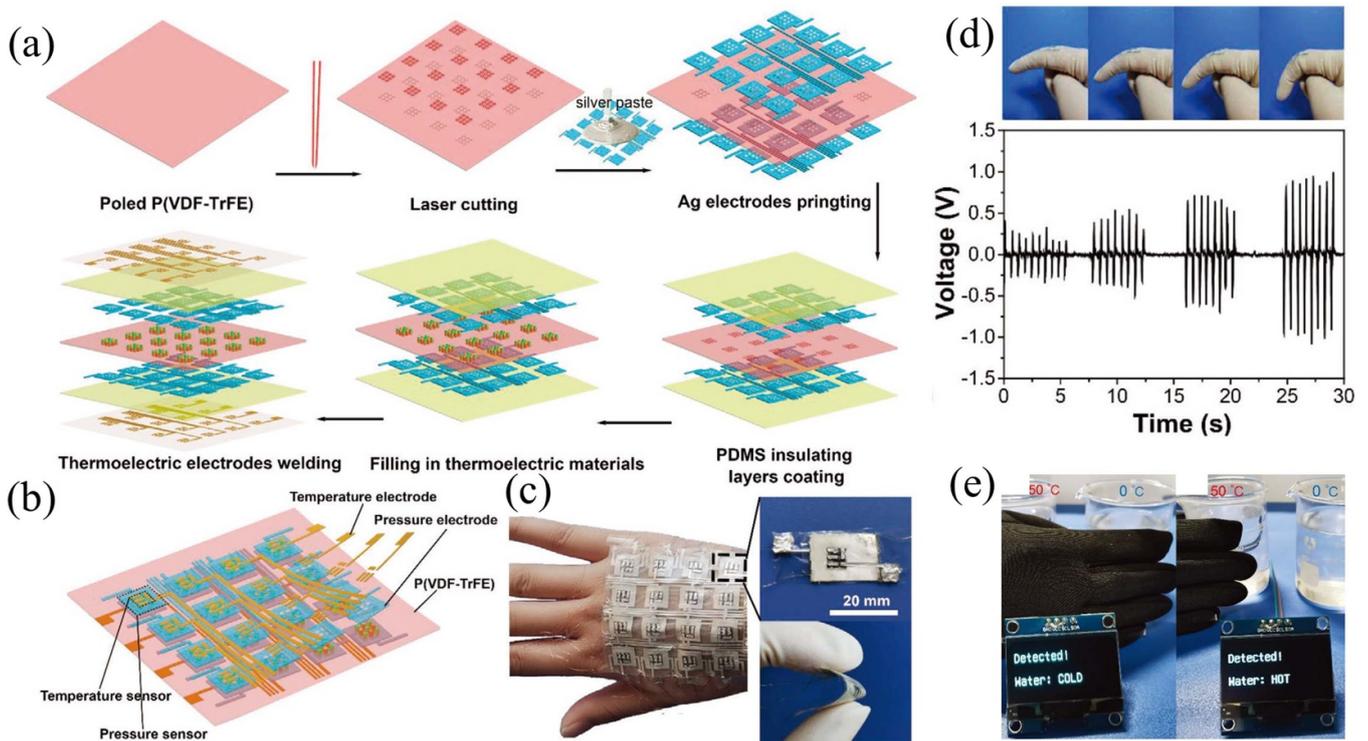


Figure 14. Weavable integrated systems. (a) Illustration of assembling a piezo/thermoelectric bimodal tactile sensor array. (b) Illustration of the structure of the integrated device. (c) Photos showing its foldability and wearability. (d) The change in voltage during the bending of the device. Sensing results when touching (e) 0 °C (left) and 50 °C (right) water by wearing the integrated device. [189] John Wiley & Sons. © 2020 Wiley-VCH GmbH.

thermoelectric materials; (3) achieving sufficient flexibility to accommodate thermal contact with different parts of the human body; (4) realizing miniaturization and compact integration of thermoelectric arms; (5) meeting the non-visual large-area heat collection requirements of thermoelectric modules; (6) ensuring stretchability and shape retention for compatibility with sustainable and stable power supply and human limb movement; (7) integrating the insulation function of fabrics with the heat transfer requirements of TEDs. Based on these requirements, future perspectives for weavable thermoelectrics can be summarized as follows (figure 15):

1. Enhancing thermoelectric performance and stability:

the future of weavable thermoelectric materials and devices necessitates continuous improvement in thermoelectric performance to enhance their usability in practical wearable applications. Researchers need to explore new weavable thermoelectric materials or enhance the weavability of existing materials to boost their thermoelectric performance while ensuring their weavability [215–221]. This may involve novel designs of compositions and micro/nanostructures and in turn, optimizing the transport of electrons and phonons [222, 223]. Additionally, weavable thermoelectric materials and devices must exhibit stability to maintain performance over extended periods of use. Also, researchers should focus on material and device optimization to minimize degradation and failure [224].

2. Improving wearability:

enhancing wearability involves several complex aspects, including the integration of weavable thermoelectric materials and devices with traditional fabrics such as clothing, ensuring that the clothing maintains adequate insulation before and after integration, portability, comfort, and more. Weavable thermoelectric materials and devices also need to be washable to a certain extent, which places higher demands on the materials and device packaging. For the washability of W-TEDs, some pioneering work has provided initial insights. For instance, to coat thermoelectric fibers with a waterproof layer, a diluted UHU adhesive solution was used to immerse the fibers and allowed to air dry [43]. This waterproof layer also serves as an electrical insulator. Additionally, we can draw inspiration from waterproof designs in other functional devices, such as incorporating polyurethane (PU) during the synthesis of CNT ink [225]. Based on the chemical structures, the amino groups within PU, which is a common component in many textiles, form hydrogen bonds between themselves. Therefore, even in water, strong adhesion between CNTs and the fabric is ensured. Alternatively, a design approach involves fully encapsulating thermoelectric fibers within waterproof and abrasion-resistant fibers, creating composite fibers, and employing advanced processes for the fabrication of these composite fibers [226]. Additionally, safety considerations are essential. For example, the components of weavable thermoelectric materials and devices should not contain

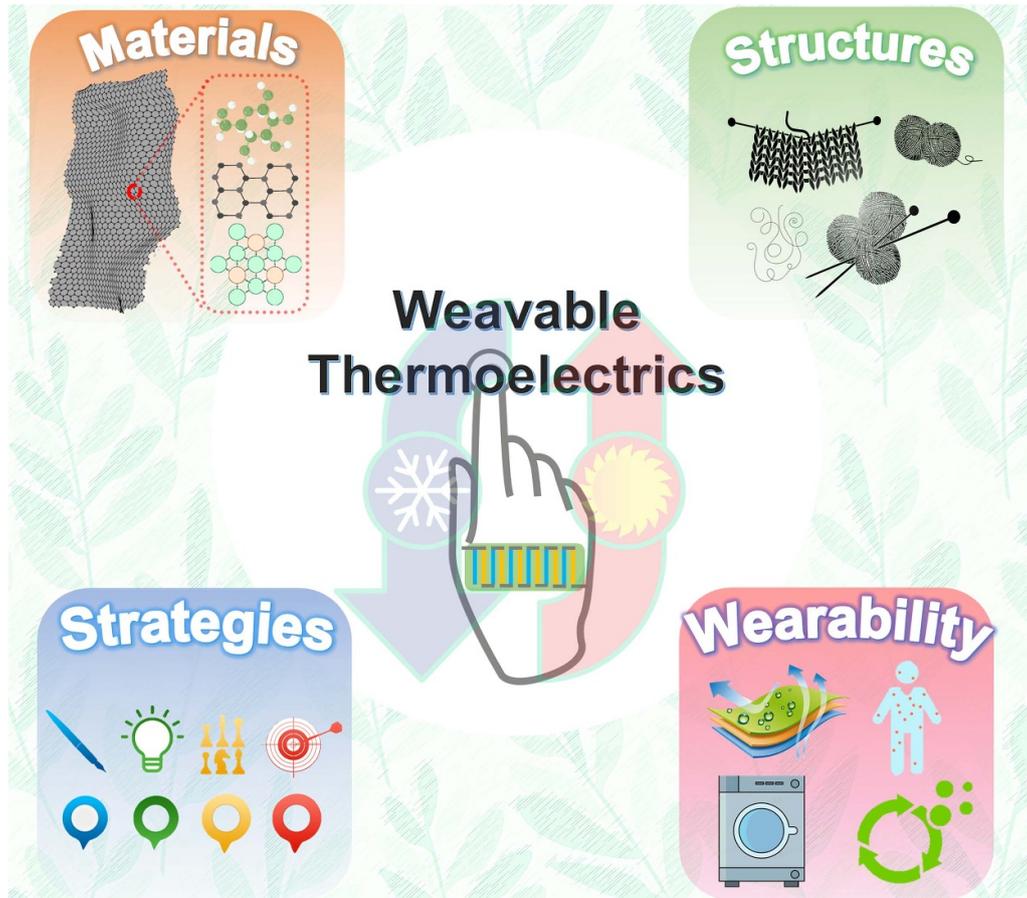


Figure 15. Illustration of outlooks for future development of weavable thermoelectrics.

highly toxic substances or allergens. The degradability and aging of weavable thermoelectric materials and devices should be considered.

3. **Heat exchanger:** for weavable TEDs, the ΔT across the thermoelectric elements is very small, expected to be less than one-tenth of the total ΔT from the body to the surrounding environment. To promote its practicality, future device designs must incorporate suitable heat exchangers. Heat exchangers play a crucial role, for instance, effectively transferring heat between the hot and cold sides of the device, thereby maintaining a stable ΔT . This contributes to improving thermoelectric efficiency. Additionally, by adjusting the design and performance of the heat exchanger, the device maintains the appropriate temperature under varying operating conditions, enhancing device performance and stability. Furthermore, heat exchangers can help TEDs adapt to different environmental conditions, as temperature variations can be significant in various application scenarios, and reduce thermal stresses on the device, extending its operational lifespan. However, for weavable thermoelectrics, heat exchangers that are small, possess certain flexibility, and offer good heat dissipation, are preferable. Traditional metal-based heat exchangers are not suitable for this purpose. Instead, new types of heat exchangers with practical potential are being explored, such as those based on phase-change materials [227, 228] or hydrogels [229, 230]. Nonetheless, heat exchangers adapted for weavable thermoelectrics are still in the early stages of research, and their adaptability requires further optimization.
4. **Scalability and large-scale manufacturing:** to achieve widespread applications of weavable thermoelectric materials and devices, it is essential to develop scalable manufacturing methods [110, 231, 232]. This includes automated production, which involves creating automated manufacturing processes to enhance preparation efficiency and reduce production costs. Furthermore, large-scale manufacturing is indispensable. Achieving large-scale manufacturing of W-TEDs is crucial to meet market demands and promote their adoption in various fields.
5. **Environmental friendliness:** future research needs to focus on the environmental friendliness of the preparation methods and materials for weavable thermoelectric materials and devices. This includes the use of green materials, which involves seeking environmentally friendly materials that can reduce production costs and environmental impact to minimize resource waste and pollution. Additionally, sustainability should be a consideration, meaning the development of sustainable manufacturing methods to reduce energy and material waste while also considering recycling and reuse.

6. **Multi-function integration:** future W-TEDs will not be limited to power generation or refrigeration but will have various integrated functions. This includes environmental monitoring, where sensors are integrated into the device to monitor environmental parameters such as temperature, humidity, gas concentrations, and more [233]. Also, health monitoring involves developing functions to monitor physiological parameters such as heart rate, body temperature, blood oxygen saturation, for personal health monitoring. Furthermore, human-machine interaction utilizes W-TEDs for human-machine interfaces, allowing machines to perceive touch, posture, force, and other information.
7. **Broader applications:** future developments will involve integrating weavable thermoelectric materials with other wearable devices to achieve broader applications [234, 235]. This includes the trending field of electronic skin [236], where W-TEDs are integrated into electronic skin to enable machines to sense the external environment and interact with it. Additionally, smart clothing involves embedding W-TEDs into garments to achieve functions such as temperature regulation and energy harvesting. Continuous research and development are ongoing for other multifunctional integrated wearable systems.

In conclusion, these directions will drive greater breakthroughs in the future applications and research of weavable thermoelectrics. Addressing these challenges may require interdisciplinary collaboration and innovation to maximize potential applications of weavable thermoelectric materials and devices.

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