Flexible Thermoelectric Materials for Wearable Energy Harvesting Engineering

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Abstract. In recent years, flexible thermoelectric (TE) materials have shown broad application prospects in the field of wearable electronics due to their unique energy conversion properties and mechanical flexibility. This article reviews the latest research progress of carbon-based, inorganic, and organic flexible TE materials, focusing on material design, performance optimization, and their applications in wearable devices. Studies have shown that through doping, compounding, and structural regulation, the Seebeck coefficient and power factor of flexible TE materials can be significantly improved, such as polyamide-doped n-type single-walled carbon nanotubes and Bi_2Te_3/PEDOT nanowire composites. The devices that is self-powered developed based on these materials have demonstrated excellent performance in the fields of human body thermal energy harvesting, temperature sensing, and health monitoring, such as high output power (18.5μW), fast response (0.6 seconds), and mechanical stability (stable performance after 1200 bending cycles). In the future, the development of flexible TE materials needs to further address the balance between conversion efficiency and wearing comfort, and promote their practical application through multidisciplinary research.

Keywords: Flexible TE materials, wearable devices, carbon nanotubes.

1. Introduction

In recent years, with the development of smart technology and information technology, more and more people have begun to pay attention to flexible wearable devices. Such devices can be integrated into clothing, worn as accessories, implanted in the human body, or printed on the skin. In terms of wearable wristband devices, global shipments reached 46.6 million units in Q1 2025, a year-on-year increase of 13%. The wearable device market has broad prospects [1]. Today, wearable devices are typically electronic devices powered by batteries. However, batteries require regular charging and replacement, hindering the device's convenience. Therefore, creating a sustainable, lightweight, self-powered generator poses a significant technical bottleneck.

Flexible thermoelectric (TE) materials are a new energy material that can convert heat and electricity. They are bendable, lightweight, and environmentally friendly, offering a solution to the challenges of wearable devices. These materials utilize the Seebeck effect to convert human body heat into electricity. Unlike photovoltaic materials, which are difficult to use at night, TE materials can operate continuously at any time. Compared to traditional generators, TE generators are less prone to low-frequency noise, effectively preventing harmful noise from affecting the human body.

TE materials which show great potential are suitable for application such as energy conversion, but their practical application is still limited by low TE conversion efficiency, which is primarily determined by the TE figure of merit (ZT value). According to the Seebeck effect, excellent TE materials must simultaneously possess a high Seebeck coefficient, high electrical conductivity, and low thermal conductivity, exhibiting the properties of a "phononic glass-electron crystal."

Since the mechanical efficiency of a person is around 15-30%, the energy a person can provide is approximately 60-180W. If the entire body is covered with TE devices with a conversion efficiency of 1%, 0.6-1.8W of power can be generated, which can power many wearable sensors [2]. However, this will sacrifice the user experience and convenience, making it difficult to satisfy the needs of practical applications. Between 2020 and 2025, the ZT value of flexible TE materials increased from 0.3 to 1.2, but the energy conversion efficiency of actual devices remained less than 2% (under human

body heat source conditions), indicating a significant gap between material performance and device engineering [3, 4]. Consequently, many researchers are focusing on improving material performance and ZT values.

This article focuses on the application of flexible TE materials in wearable devices, specifically examining a range of the latest flexible TE wearable devices. These include self-repairing and recyclable flexible TE devices, temperature sensors, and flexible strain sensors. This article aims to provide researchers with more research directions and make TE research more cost-effective.

2. Flexible TE MATERIALS

2.1. Carbon-Based Flexible TE Materials

2.1.1 CNT Flexible TE Materials

CNTs are generally classified into single-walled carbon nanotubes(SWCNTs) and multi-walled carbon nanotubes. SWCNTs have a unique electronic structure, high electrical conductivity, and processability, but are limited by their inherent high thermal conductivity. In recent years, Liu's team has achieved a room-temperature power factor of 16.39 μW cm-1K-2 for SWCNT/Cu2Se composite films by cold pressing with Cu2Se, with a quality factor increased by 500% [5]. This also demonstrates the enormous potential of inorganic composites.

Typically, the construction of wearable TE Generators (TEGs) requires p-type and n-type flexible TE materials, which are thermally connected in series and parallel. However, the progress of n-type flexible TE materials lags behind that of p-type materials, because their sensitivity is unstable for the environment. The application of wearable TEG is inevitably hindered by this limitation. CNT fibers or yarns have become one of the most promising flexible n-type TE materials because they have high conductivity, excellent flexibility, and can be converted from p-type to n-type by doping easily. Khong et al. used aerosol to dope SWCNT. The results showed that compared with traditional liquid-phase doping, it is easier to control the nanotube morphology, and there is no residual surfactant that increases the junction resistance of the nanotubes and induces defects. They used aerosol chemical vapor deposition to prepare SWCNT films, using the Budua reaction on an iron-based catalyst [6]. This makes the preparation of SWCNTs more adjustable and precisely controlled.

By using three poly (Fe-CPT) derivatives (Fe-P1, Fe-P2, Fe-P3) containing ferrocene and thiophene functional groups, the polymer was composited with SWCNT to prepare a flexible TE film. Sun et al. characterized the structure, morphology, and thermal stability of the materials through experiments, and calculated and analyzed the interaction and electronic structure between the polymer and SWCNT [7]. The results showed that Fe-P3 had the highest binding energy with SWCNT (63.36 kJ mol^{-1}), and electron transfer significantly reduced the HOMO-LUMO energy gap, improved the conductivity, and the OTEG module based on Fe-P3/SWCNT had an output power of 1.18 μW at a temperature difference of 72 K, which was three times that of the other two materials [7]. This study generates a new research direction for organic TE materials and highlights the potential of organic TE technology.

2.1.2 Graphene Flexible TE Materials

Graphene has excellent electrical, thermal conductivity, adjustable nanostructure, mechanical flexibility, chemical stability, and a large specific surface area, which has shown great potential in TE materials. Graphene nanosheets are popular on the market, accounting for more than 80% of the total market sales. This is because graphene nanosheets have higher mechanical strength and lower production costs, which is different from single-layer graphene. However, the lower electrical conductivity of graphene nanosheets is not conducive to TE applications [8].

The Uchida team enhanced the TE properties of graphene films through π-π interactions. They used spin coating to prepare a graphene/PEDOT: PSS composite. The aggregation of PEDOT: PSS on graphene films through π-π interactions was confirmed by characterization methods such as atomic force microscopy, X-ray diffraction, and Raman spectroscopy. The experimental results showed that

the power factor ($S^2\sigma$) of this film was 1.6 times higher than that of pure graphene film, reaching 1.2 μW cm^{-1} K^{-2} [9]. This study shows that π-π interactions can significantly improve TE performance and generate a valuable strategy for the development of high-performance, low-cost flexible TE films.

2.2. Inorganic Flexible TE Materials

2.2.1. Ag-S-Based Flexible TE Materials

Silver-based chalcogenides (Ag_2Q, Q = S, Se, Te) have become ideal materials for flexible TE generators (TEGs) because of their incredible near-room-temperature TE properties and unique flexibility. Among them, Ag_2Se thin films have a ZT value of up to 1.2 at 300K, combining high conductivity and bending adaptability. Their devices can output a power density of 35.5 W/m² when the difference of temperature is 30K; Ag_2S achieves metallic-level ductility (compressive strain > 50%) through Se/Te alloying and dual-phase engineering; while nanostructured Ag_2Te exhibits a ZT value of 1.37, it is limited by the scarcity of Te [10]. A specific comparison is shown in Table 1. AgCuSe induces a "brittle-to-flexible" transition through S doping, filling the gap in p-type flexible materials. Current challenges include Ag_2S doping restrictions, difficulties in preparing p-type Ag_2Se, and Te resource bottlenecks. In the future, new preparation technologies, self-healing materials, and optimized device designs need to be developed to promote their application in wearable electronic self-powered systems.

Table 1. Comparison of Ag-S-based materials

Material	Maximum ZT value (temperature)	Features	Application potential
Ag_2Se film	1.2 (300 K)	High conductivity, excellent flexibility	Wearable device power supply
Ag_2Te nanomaterials	1.37 (373 K)	Nanostructure-enhanced phonon scattering	Special fields (such as aerospace)
AgCuSe	1.6 (670 K)	"Brittle-to-flexible" transition, p-type conduction	Flexible TE devices

In order to further improve the ZT value and prove that vertical TEG can make better use of human body temperature to generate electricity, Won et al. doped flexible free-standing Ag_2Se thin films through Cu_2Se nanoparticles. The ZT of the Ag_2Se thin film that uses only 50 ppm of Cu_2Se at room temperature was increased from 0.46 to 0.55. Moreover, this still maintains its ultra-low bending radius of 0.4 mm [11]. Unlike most thin-film flexible TE devices that are in-plane modules, they designed a vertical TEG, proved the possibility of vertical TEG in the wearable field, and generated a new strategy for high-performance wearable TEs.

To retain the TE conversion capability of inorganic TE materials and significantly improve their flexibility, Wang's team, based on the functional element sequence strategy, constructed a "core-shell" structure (S-doped Ag_2Se as the core, Se-doped Ag_2S and amorphous S as the shell) and introduced polymers (PVP or PPy). Among them, the power factor of the S-doped Ag_2Se film reached 954 $\mu W \cdot m^{-1} \cdot K^{-2}$, and the conductivity remained at 94.4% after bending 2000 times; Ag_2Se/PVP reached 1910 $\mu W \cdot m^{-1} \cdot K^{-2}$, with better flexibility; Ag_2Se/Se/PPy reached 2240 $\mu W \cdot m^{-1} \cdot K^{-2}$, and had excellent mechanical stability [12]. This study successfully achieved the unity of high performance and high flexibility of Ag_2Se-based TE films through functional element sequence design, providing new ideas for flexible TE technology.

2.2.2 Cu2Se Flexible TE Material

Cu2Se has high carrier mobility and excellent ZT value, and has been widely used in rigid TE devices. Its unique "liquid-like" properties (Cu^+ ions can migrate freely in the lattice) cause it to undergo a reversible phase transition (α→β phase) around 400 K, significantly affecting the electrothermal transport performance [13]. However, considering the inherent brittleness of bulk inorganic materials, how to give them a certain degree of flexibility is a difficult problem. In the past

few years, processing inorganic TE materials into two-dimensional thin films has become an effective strategy.

Chen et al. used a sulfur (S) doping strategy to optimize the TE properties of β-Cu_2Se thin films and improve the output power of the corresponding flexible TE devices. The study prepared β-$Cu_2(Se,S)$ solid solution films with different S doping concentrations by magnetron sputtering technology. The experimental results showed that S successfully replaced Se into the lattice, forming a uniform solid solution structure. As the S content increased to a maximum of 6.3 at.%, the carrier concentration gradually decreased, while the mobility increased, resulting in a decrease in conductivity but a significant increase in the Seebeck coefficient, reaching a maximum of 57 μV K^{-1} at room temperature [14]. This study broke through the limitations of Cu2Se's electronic structure, transport properties, and relatively low TE conversion efficiency, and provided a basis for effective temperature sensors with fast response characteristics.

In order to improve the Seebeck coefficient, Wang's team prepared flexible $Cu_2Se_{1-x}I_x$ (x=0, 0.02, 0.04, 0.06) TE films by a hydrothermal method combined with vacuum filtration and hot pressing. The study found that iodine doping as n-type doping provided additional electrons, reduced the carrier concentration, resulting in a decrease in conductivity but a significant increase in the Seebeck coefficient. When x=0.04, the film achieved the optimal power factor (~566.9 μW m^{-1} K^{-2}) at room temperature as well as excellent flexibility [15]. The six-legged TE device based on $Cu_2Se_{0.96}I_{0.04}$ film, achieving a maximum output power of 1.41 μW and a power density of 4.02 W m^{-2} when the ΔT=22.8 K proposed an effective strategy for high-performance flexible TE materials [15].

3. Application of Flexible TE Materials in Wearable Fields

3.1. Wearable TE Devices Based on Organic Flexible TE Materials

Organic flexible TE materials have attracted much attention in the fields of wearable energy harvesting and personalized thermal management due to their combined TE conversion properties and flexibility. Traditional inorganic TE materials (such as Bi_2Te_3) are highly efficient but also very rigid, while organic materials (such as conductive polymers) make up for this shortcoming through flexibility, low thermal conductivity, and solution processing advantages. Zhu's team successfully developed a revolutionary self-healing and recyclable flexible TE device by combining a dynamic covalent polyimide substrate, liquid metal electrodes, and Bi_2Te_3/Sb_2Te_3 TE legs. The device achieves excellent self-healing ability (24-hour repair, performance recovery >90%) and full recyclability (performance loss <9%) thanks to the dynamic covalent network, while also being able to fit tightly to human skin. After integrating the PID control system, the body temperature can be dynamically adjusted to a comfortable range, and local cooling treatment for fever or sprains (minimum 18.2°C) can be achieved. In addition, the body heat energy can be collected through the boost circuit to drive the LED, providing a high-performance and sustainable solution for wearable energy and personalized thermal management, as shown in Figure 1 [16].

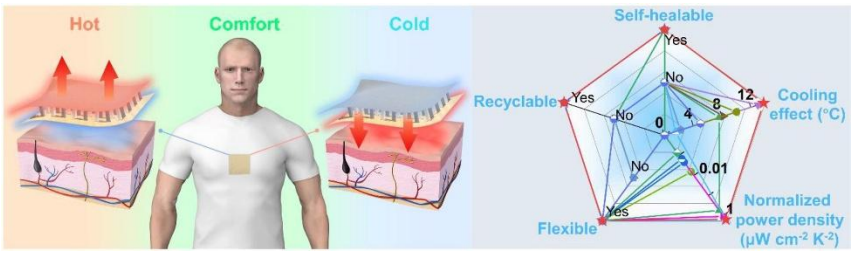

Figure 1. Self-repairing and recyclable flexible TE device [17]

Du et al. fabricated an ultrathin wearable TE patch based on an organic ion gel electrolyte, which has excellent performance and application prospects. The patch uses a porous polymer skeleton with an "onion epidermal cell" structure to support an organic ion gel electrolyte.It achieves a ZT value improvement to 900% and exhibits a high Seebeck coefficient of 28mV K^{-1} and an energy conversion

efficiency of 1.3%. Its ultrathin (0.197mm), flexible, and biocompatible design enables it to fit closely to the skin, enabling the collection and conversion of human thermal energy [18]. This makes the patch show a wide range of application potential in the fields of thermal charging energy storage, material identification, contact/non-contact temperature detection, and photothermal conversion.

3.2. Wearable TE Devices Based on Inorganic Flexible TE Materials

Inorganic flexible TE materials generally have better TE performance than organic flexible TE materials. However, due to their rigidity, they are difficult to make into flexible devices. They usually need to be combined with organic materials or carbon-based materials to make up for this defect. Du's team successfully prepared high-performance Bi_2Te_3/PEDOT nanowire composite films using thermal deposition and thermal shock techniques. Based on this material, they further developed a flexible cross-plane TE generator (TEG). The device consists of 16 TE legs connected by liquid metal ($Ga_{78}In_{225}$) and encapsulated in PDMS, which is both flexible and stable. When the temperature difference is 17.8K, its output power density can reach 3.48 μW cm^{-2}, and it is expected to be further increased to 70.6μW cm^{-2} at a temperature difference of 80K. The overall performance is significantly better than most organic and organic/inorganic hybrid TE materials [19]. The sensor array has excellent flexibility and mechanical stability, can conform to curved surfaces (such as human skin), and is suitable for temperature monitoring and energy harvesting applications in wearable electronic devices. These characteristics give it broad application potential in fields such as health monitoring, human-computer interaction, and high-temperature protection.

A sensitive and flexible piezoresistive strain sensor based on a tellurium (Te) nanomesh structure is fabricated by Wei et al.. They prepared a Te nanomesh structure on a flexible polyimide substrate by physical vapor deposition. This unique mesh structure gives the sensor extremely high strain sensitivity, with a gauge factor of up to 9.93×108. After the sensor was treated with polydimethylsiloxane coating, its stability was significantly improved, and its performance remained stable even after 1000 load-release cycles. In addition, they also researched a system that is wireless and real-time based on the Arduino platform, further verifying its potential in wearable device, which provides new ideas for the design of high-performance flexible strain sensors and promotes the development of related technologies [17].

3.3. Wearable TE Devices Based on Carbon-Based Flexible TE Materials

In recent years, flexible TE materials have shown great application potential in the field of wearable electronics, providing innovative solutions for self-powered sensing and energy harvesting. Xiao et al. innovatively used the amide group in polyamide (PA6/PA66) as an electron donor to successfully convert p-type SWCNTs into n-type TE materials, and prepared a flexible TE module based on this. The device, consisting of five p-n junctions in series, can output a thermovoltage of 43.1 mV and a power of 18.5 μW at a temperature difference of 80 K, and can respond to temperature changes quickly (0.6 seconds), realizing self-powered human activity monitoring and temperature communication. The device also exhibits excellent mechanical durability, and its performance remains stable after 1200 bends [20]. This study provides a novel n-type doping strategy and a feasible material solution for the development of efficient, stable, and multifunctional wearable self-powered systems.

The KIM team used a wet spinning process to prepare graphene oxide fibers, and used a selective chemical reduction strategy to differentially reduce different sections of the fiber by regulating the concentration of hydroiodic acid solution, thereby constructing a graphene composite fiber with a seamless connection structure. One part of the fiber is mildly reduced graphene oxide and the other part is highly reduced graphene oxide. Due to the different reduction degrees of the two parts, their Seebeck coefficients are different, which gives the fiber thermocouple function. Experiments show that the graphene thermocouple has high sensitivity, good linearity and fast response characteristics in the range of room temperature to 70°C [21]. In addition, the fiber exhibits excellent mechanical flexibility and stability and can be woven into cotton gloves to realize wearable temperature sensing

applications. This study generates a new idea for the development of high-performance flexible temperature sensors.

4. Conclusion

Research on flexible TE materials in the field of wearable devices has achieved important breakthroughs. Through material innovation and device design, they have successfully achieved the combination of efficient TE conversion and mechanical flexibility. Carbon-based materials such as single-walled carbon nanotubes and graphene have shown significant advantages due to their unique electronic structure and lightweight properties; inorganic materials such as Ag_2Se and Cu_2Se have significantly improved the output performance of flexible devices through composite and doping strategies; organic materials have expanded their application scenarios with their dynamic covalent bond design and self-healing functions. Studies have shown that optimized flexible TE devices have demonstrated excellent performance in areas such as human body thermal energy harvesting, temperature sensing, and health monitoring, including high output power, fast response, and excellent mechanical stability. These advances have laid a solid foundation for the development of wearable electronic devices.

Although flexible TE materials have achieved remarkable results, they still face challenges such as performance bottlenecks, environmental stability, and integration technology. Future research needs to further explore new composite material systems and break through the ZT value limitation through strategies such as heterogeneous structure design and quantum dot modification. At the same time, it is crucial to combine artificial intelligence to optimize the device structure and improve the balance between energy collection efficiency and wearing comfort. In addition, the development of biocompatible materials and promoting their integration with self-powered systems will expand the application potential of flexible TE materials in fields such as medical monitoring and personalized health management. Interdisciplinary cooperation and engineering breakthroughs will be the key to realizing the commercial application of flexible TE technology and providing sustainable energy solutions for the next generation of wearable electronics.

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