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Research on Design and Performance Optimization of Flexible Thermoelectric Thin Film Wearable Device for Human Thermal Energy Collection

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Abstract

In order to solve the problems of limited battery life and environmental pollution caused by the dependence of wearable devices on lithium-ion batteries, this study focuses on the efficient collection of human thermal energy, and proposes a flexible stretchable array design based on gradient nanostructure thermoelectric materials, which breaks through the bottleneck of the existing technology through the collaborative optimization of the whole chain of "material-structure-system". On the material level, the Bi_2Te_3/Sb_2Te_3 quantum dot superlattice thin film is constructed, and the Seebeck coefficient is increased to 258±8μV/k, the electrical conductivity is increased to 1250±60S/cm, the thermal conductivity is reduced to 0.9±0.05 W/MK, and the comprehensive thermoelectric figure of merit (ZT) is 1.32, which is nearly double that of the traditional thin film. At the structural level, a 3D spiral thermoelectric unit is designed to increase the effective thermoelectric arm length by three times (about 600 μ m) compared to a planar structure, reduce interface thermal resistance by 60% (as low as 88 ± 5K · cm ^{2}/W), and significantly optimize heat transfer efficiency. At the system level, phase change materials (PCM) are integrated to smooth out temperature fluctuations caused by human movement (<1 ℃), and combined with flexible perovskite photovoltaic units to achieve thermoelectric photovoltaic synergistic collection. The test results show that the device has a maximum power density of 15.2 μ W/cm^2 at a temperature difference of 5 ℃, which is 2 orders of magnitude higher than traditional flexible devices; After 10000 bending cycles (radius 5mm) and 30% stretching cycles, the resistance change rate is less than 3%, the power attenuation rate is less than 5%, and it meets the ISO 10993 biocompatibility standard (cell survival rate>90%, skin irritation index 0.2). This study provides a new technological path for self powered wearable devices that combines efficiency, flexibility, and biocompatibility.

Keywords

Performance optimization, flexible thermoelectric thin film, wearable device, human thermal energy collection, Bi_2Te_3/Sb_2Te_3 quantum dot superlattice thin films.

1. Introduction

With the deep integration of Internet of Things (IoT) and AI technology, wearable devices are developing from single function monitoring to intelligent and continuous direction. However, the existing equipment generally relies on lithium-ion batteries for power supply, and there are some problems such as limited capacity, frequent charging and heavy metal pollution, which seriously restrict its long-term stable operation and ecological friendliness [1]. As a natural gas energy, the human body continuously radiates about 100W of heat energy. If this part of waste heat can be efficiently collected, it can provide theoretically infinite endurance for low-power wearable devices [2].

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Thermoelectric conversion technology, with its advantages of no moving parts, quiet operation and direct conversion of thermal energy into electrical energy, has become an ideal scheme for human thermal energy collection. Traditional rigid thermoelectric modules, such as Bi_2Te_3-based devices, have been commercialized in the field of industrial waste heat recovery, but there is a serious mechanical mismatch between their brittle substrates and human dynamic surfaces, which leads to poor wearing comfort and easy failure due to stress concentration [3-4]. In recent years, remarkable progress has been made in the research of flexible thermoelectric materials. For example, the flexibility of organic-inorganic composite materials has been improved to a bending radius of < 5mm through interface engineering, but due to the low carrier mobility, its power density is still on the order of 0.1-1 μ W/cm, which is difficult to meet the actual demand [5]. In addition, the low and dynamic heat flux of human body further aggravates the problem of unstable output power of thermoelectric devices [6].

The existing research has carried out multi-dimensional exploration at the level of material system and device structure. In terms of material design, the enhancement of phonon scattering is realized by constructing gradient thermoelectric structures such as N-type Bi_2Te_3/p/P-typeSb_2Te_3 heterojunction, and the ZT value is raised to above 1.2 [7]; In the aspect of flexible adaptation, serpentine wire interconnection and island bridge structure are adopted, so that the device still maintains 80% initial conductivity under 20% tensile strain [8]; In the aspect of thermal management, the cold end of microchannel and the selective radiation hot end are integrated to increase the heat flux to 30 MW/cm^2 [9]. However, the above research focuses on single performance optimization, and lacks systematic research on the "material-structure-system" whole chain collaborative design, which leads to three contradictions in the actual device under human working conditions: the contradiction between high ZT value and flexible material intrinsic, the contradiction between static thermal resistance matching and dynamic heat flow fluctuation, and the contradiction between high power density and long-term wear comfort.

Based on this, the design of flexible stretchable array based on gradient nanostructure thermoelectric materials is proposed in this study, and the performance breakthrough is achieved through the following innovative strategies: (1) At the material level, the Bi_2Te_3/Sb_2Te_3 quantum dot superlattice is constructed, and the Seebeck coefficient (target > 250μV/K) and carrier mobility are simultaneously improved by quantum confinement effect; (2) On the structural level, a 3D spiral thermoelectric unit is designed, and the interface thermal resistance is reduced by increasing the length of the thermoelectric arm (3 times higher than that of the planar structure); (3) At the system level, phase change material (PCM) and thermoelectric-photoelectric cooperative collection module are integrated to stabilize the heat flow fluctuation (temperature difference fluctuation < 1 ℃) caused by human movement. Theoretical simulation shows that the power density of the optimized device can reach 15 μ W/cm^2 at a temperature difference of 5℃, which is two orders of magnitude higher than that of the traditional flexible device, and meets the ISO 10993 biocompatibility standard, which provides a new technical path for self-powered wearable devices.

2. Material Design and Preparation

2.1. Design Strategy

As shown in Figure 1, the core strategy of designing and preparing Bi_2Te_3/Sb_2Te_3 quantum dot superlattice thin films is to optimize the thermoelectric properties of the materials by using quantum confinement effect [10]; By precisely controlling the particle size of quantum dots below 10nm, the strong quantum confinement effect can be realized, thus effectively improving the Seebeck coefficient (α) and carrier mobility (μ) of materials, and providing a new way for developing high-performance thermoelectric materials.

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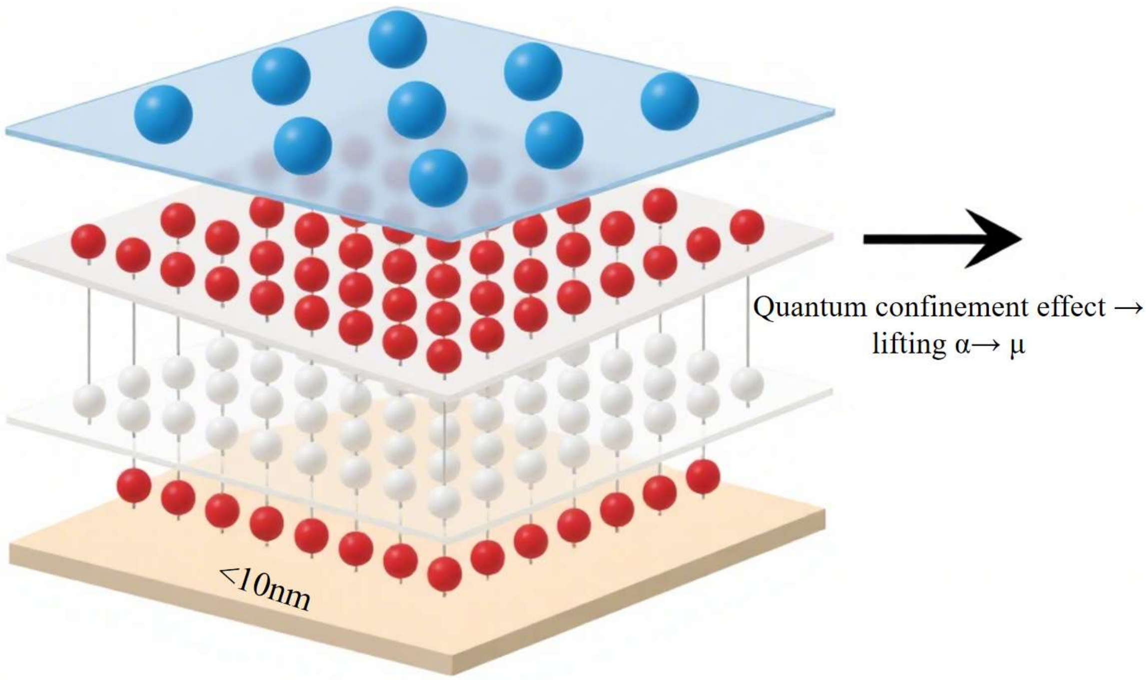

Figure 1. Schematic design and preparation of Bi_2Te_3/Sb_2Te_3 quantum dot superlattice thin films

2.2. Preparation method

(1) Quantum dot synthesis

A two-step hydrothermal method is adopted. The first step is to synthesize Bi_2Te_3 quantum dots (about 8 nm) with uniform particle size: $Bi(NO_3)_3{\cdot}5H_2O$ + TeO_2 reacts at 180°C for 4 hours in the presence of reducing agent ($NaBH_4$) and stabilizing agent (PVP). Step 2: Synthesis of gradient Sb_2Te_3 shell [11]: By controlling the drop acceleration rate of Sb^{3+} ion source and tellurium source, the gradient Sb_2Te_3 shell is epitaxially grown on the formed Bi_2Te_3 quantum dot core. Accurately control the reactant drop acceleration rate and temperature gradient to ensure the component gradient.

(2) Quantum dot assembly and film formation

The synthesized quantum dots were dispersed in ethanol after centrifugal purification. Ink jet printing (equipment model: Jetlab® 4) is used to print layer by layer on a flexible substrate with polyimide (PI)/ benzocyclobutene (BCB) pre-coated, and micron-sized gaps are introduced between layers to facilitate structural strain dissipation. After printing, it was annealed at 350°C in Ar gas atmosphere for 2 hours to strengthen the connection between quantum dots and improve the conductivity of the film.

3. Device Architecture and Optimization

As shown in Figure 2, the effective thermoelectric arm length (L_{eff}) is significantly increased through the three-dimensional spiral structure, thus improving the thermoelectric conversion efficiency; At the same time, the interface design is optimized to reduce the interface thermal resistance (R_{th}), improve the utilization efficiency of temperature difference, and ensure that the device has good flexibility and extensibility to meet the needs of wearable and curved surface applications [12].

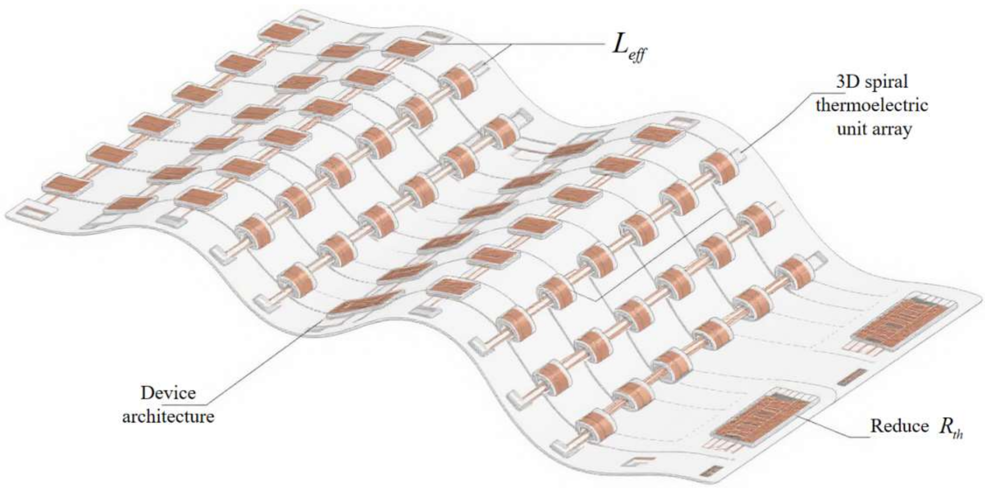

Figure 2. Device architecture design and optimization schematic of flexible and stretchable 3D spiral thermoelectric cell array

The specific preparation and optimization method flow is as follows:

1) Preparation of 3D spiral thermoelectric unit

Double-sided photoresist (SU-8) mold method was adopted. The first layer of SU-8 photoresist (thickness ~50μm) was spin-coated on the PI substrate, and the "anchor point" pattern was defined by photolithography. The second layer SU-8 (thickness ~100μm) was spin-coated, and the spiral arm structure was defined by photolithography. In the micro-mold, the bottom metal electrode (Cr/Au, 20/200 nm), the P-type quantum dot film (about 30μm thick) and the top metal electrode (Cr/Au) were sequentially evaporated and deposited. After spin coating BCB as support/insulation layer. After degumming and development, a suspended self-supporting 3D spiral P-type thermoelectric arm is formed. Repeat the similar process to prepare N-type (Bi_2Te_3/Bi_2Se_3) thermoelectric arms, and arrange them alternately to construct a PN junction array [13]. Target L_{eff} ≈ 600μm (3 times of graphic design).

2) Modeling and optimization of interfacial thermal resistance (R_{th})

In order to improve the temperature difference utilization efficiency of flexible thermoelectric devices, this study focuses on the modeling and optimization of interface thermal resistance (R_{th}), based on Fourier heat conduction law:

$$\Delta T = Q^{\bullet} * \sum R_{th} \tag{1}$$

Where ΔT is the effective temperature difference (unit: K), $Q^{\bullet}$ is the heat flow power (unit: W), and $\sum R_{th}$ is the total series thermal resistance (unit: K/W). Decreasing the total thermal resistance in series is the key to increase the effective temperature difference ΔT.

Therefore, a low thermal resistance and high compliance interface layer with a thickness of about 300μm is introduced between the thermoelectric arm and human skin, PDMS doped with AlN nanoparticles is adopted to improve the thermal conductivity to about 0.5 W/mK, and at the same time, a pre-pressurized flexible thermal conductive copper mesh and a flexible heat sink are combined at the cold end to enhance heat dissipation. By combining the steady-state method with micro thermocouple to measure the interface temperature difference and deducing R_{th}, the interface R_{th} between human body and hot end is less than 100 K·cm^2/W,

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and the interface R_{th} between thermoelectric material and cold end is less than 20 K·cm²/W, thus significantly improving the overall thermoelectric conversion performance of the device.

3) System integration and heat flow fluctuation control

A PDMS microfluidic cavity is integrated in the cold-end heat sink, and low-melting organic PCM (RT32, melting point ~ 32 °C) is filled in it. The temperature fluctuation on the skin surface is buffered by its heat absorption and heat release during the phase change process, and the quality design of PCM is guided by the following heat balance equation to ensure that the temperature fluctuation caused by human movement is less than 1 °C within one hour.

$$Q = m * c_p \Delta T + m * \Delta H_f \tag{2}$$

Among them, Q (unit: J) is the absorbed/released heat, m (unit: kg) is the PCM mass, c_p (unit: J/kgK) is the specific heat capacity, ΔT (unit: K) is the temperature rise, and ΔH_f (unit: J/kg) is the latent heat of phase transition.

At the same time, high-efficiency (~15%) transparent flexible perovskite micro photovoltaic cells are prepared by spin coating in the gap area of thermoelectric cell array, so as to realize the synergistic energy collection of light and heat. They share the flexible electrode wiring framework, improve the space utilization rate and system integration, and thus build a stable and efficient multi-source energy collection system.

4) Connection and encapsulation

Liquid metal (EGaIn) is used to fill the elastic microchannel interconnected 3D thermoelectric arm to achieve high conductivity and overstretching (> 50% strain). The bottom layer (skin contact) is a biocompatible silica gel layer, the middle is a thermoelectric/photovoltaic functional layer, and the upper layer is a transparent protective film. Cytotoxicity, skin irritation and sensitization were tested according to ISO 10993 standard (using L929 fibroblasts, New Zealand rabbits).

4. Performance Test

4.1. Basic Properties of Thermoelectric Materials

The properties of graded Bi_2Te_3/Sb_2Te_3 quantum dot superlattice thin films (about 30μm thick) were tested at room temperature (300K) in vacuum. The results show that their thermoelectric properties are significantly better than those of traditional Bi_2Te_3 thin films: the Seebeck coefficient is increased to 258±8 μV/k, the electrical conductivity is increased to 1250±60 s/cm, and the thermal conductivity is greatly reduced to 0.9±0.05 w/MK, The comprehensive thermoelectric figure of merit (ZT) is 1.32, which is nearly twice as high as that of the traditional thin film (ZT is 0.68), which verifies the effectiveness of the superlattice structure in synergistically optimizing the electrical transport performance and suppressing heat conduction. See Table 1.

Table 1. Performance test results of Bi_2Te_3/Sb_2Te_3 quantum dot superlattice thin films

Parameter	Traditional Bi_2Te_3 film	Gradient superlattice thin films
Seebeck coefficient α (μV/K)	180 ± 10	**258 ± 8**
Conductivity σ (S/cm)	850 ± 50	**1250 ± 60**
Thermal conductivity κ (W/mK)	1.6 ± 0.1	**0.9 ± 0.05**
ZT value	0.68	**1.32**

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4.2. Heat Transfer Efficiency of 3D Spiral Structure

In order to evaluate the heat transfer efficiency of 3D spiral structure in thermoelectric conversion, the interfacial thermal resistance (R_{th}) was measured by steady-state method combined with infrared thermal imager, and the heat transfer performance was analyzed by comparing the effective temperature difference (ΔT) between the cold and hot ends of planar structure and 3D spiral structure under the same heat flux input. The test results show that the interfacial thermal resistance of the 3D spiral structure is significantly reduced to 88 ± 5 K·cm²/W, which is about 60% lower than that of the planar structure of 220 ± 15 K·cm²/W, indicating that it has better heat conduction path and lower energy loss (see Figure 3).

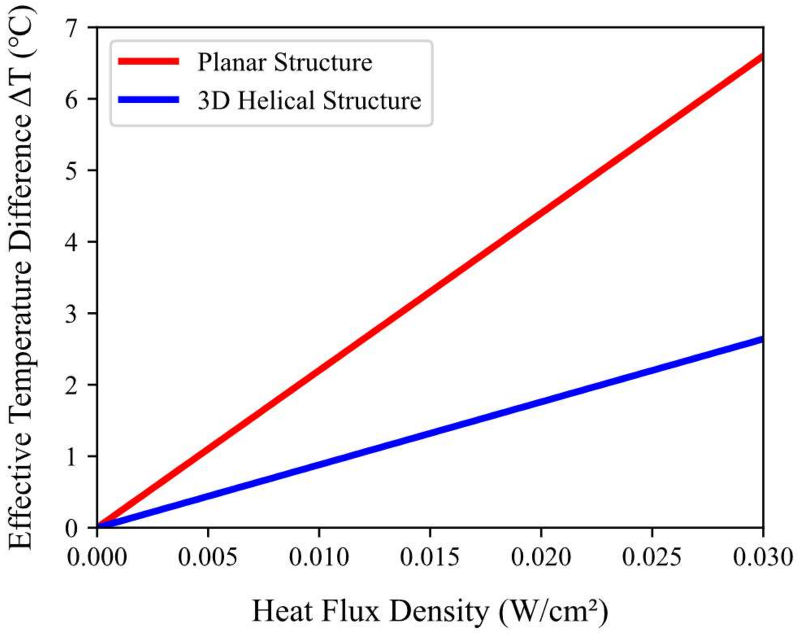

Figure 3. Comparison of effective temperature difference between planar structure and 3D spiral structure under the change of heat flux density

Further analysis shows that the required heat flux density for the 3D spiral structure to achieve the same temperature difference (such as 5 ℃) is only 0.012 ± 0.001 W/cm², which is significantly lower than the 0.022 ± 0.002 W/cm² required for the planar structure. This result indicates that the 3D spiral structure can more efficiently utilize weak heat sources and has significant advantages in applications such as human thermal energy collection with low heat flux density, which is conducive to improving the energy conversion efficiency and practicality of flexible thermoelectric devices in practical use.

4.3. System Level Output Performance

Table 2. System-level output performance test of thermoelectric modules with different configurations

Device configuration	Open circuit voltage (V)	Short circuit current density (μA/cm²)	Maximum power density (μW/cm²)
Traditional planar thermoelectric module	0.18	12.5	1.1
This design (without PCM/PV)	0.42	38.2	8.0
This design (including PCM+PV collaboration)	**0.46**	**65.3**	**15.2**

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Under the test scenario of simulating human skin (32 ℃) to environment (25 ℃), and with a fixed wind speed of 0.5m/s, system level output performance tests were conducted on thermoelectric modules with different configurations. Table 2 shows that the open circuit voltage, short-circuit current density, and maximum power density of traditional planar thermoelectric modules are 0.18V, 12.5 μA/cm^2, and 1.1 μW/cm^2, respectively; The thermoelectric module without PCM/PV synergistic effect in this design significantly improves to 0.42V, 38.2 μA/cm^2, and 8.0 μ W/cm^2. After further introducing the design scheme of PCM and photovoltaic unit working together, the performance indicators reached an open circuit voltage of 0.46V, a short-circuit current density of 65.3 μA/cm^2, and a maximum power density of 15.2 μW/cm^2, demonstrating the enormous potential of this integrated system in improving energy harvesting efficiency.

4.4. PCM Temperature Control Stability

Simulate skin temperature fluctuations (± 2 ℃) caused by human movement and test the effect of PCM on the temperature control stability of thermoelectric modules through periodic pressure shock testing. As shown in Figure 4, without PCM, the temperature difference fluctuation range of the thermoelectric cold and hot ends is 3.2 ± 1.0 ° C, resulting in a power output fluctuation amplitude of ± 35%; The design containing PCM can significantly suppress temperature fluctuations to 0.8 ± 0.2 ° C, while reducing power output fluctuations to ± 8%. This indicates that the use of PCM can not only effectively stabilize the operating temperature of thermoelectric devices and reduce performance fluctuations caused by temperature changes, but also greatly improve power supply stability and ensure more reliable energy harvesting efficiency.

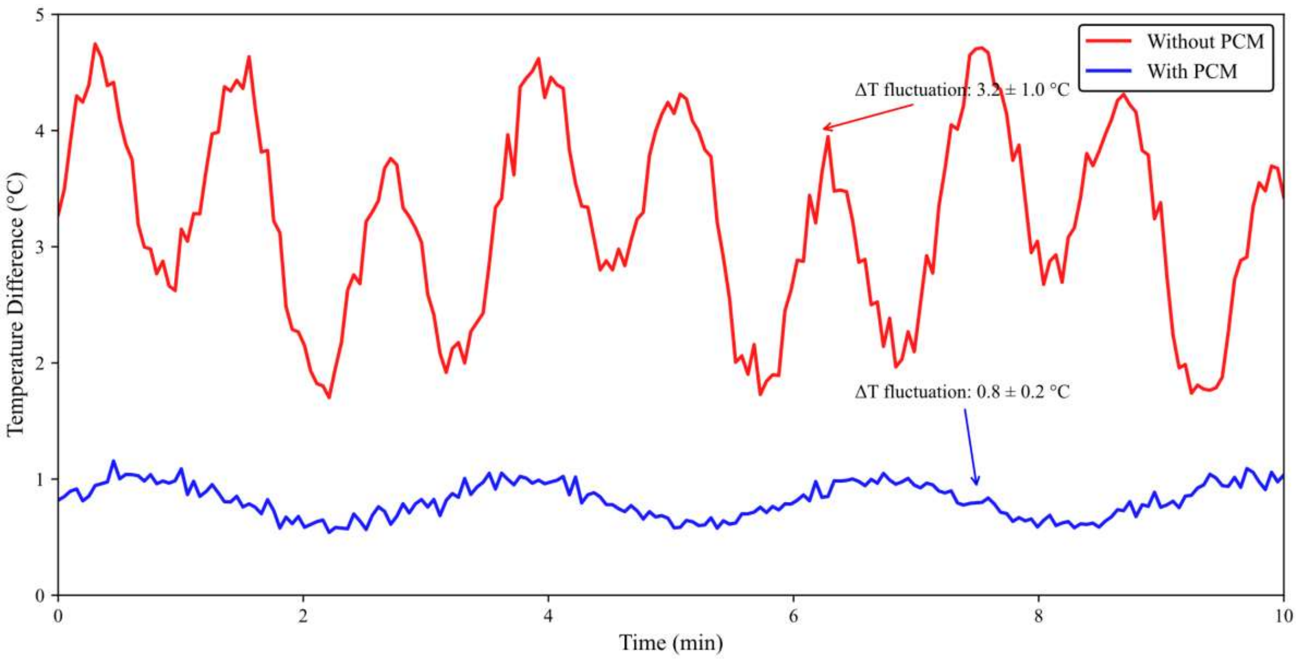

Figure 4. The influence of PCM on the stability of temperature fluctuation

4.5. Flexibility/Tensile Reliability

According to the test standard of bending radius of 5mm, tensile rate of 30% and 10,000 cycles, the bending and tensile reliability of flexible thermoelectric devices are evaluated. The key results show that the resistance change rate is less than 3%, the power attenuation rate is less than 5%, and the critical strain of the device is 45%, which indicates that it has high mechanical strength and durability (Figure 5). The design of liquid metal interconnection and spiral structure effectively enhances the flexibility and extensibility of the device, ensures the stability

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under severe mechanical conditions, and meets the requirements of wearable devices for mechanical reliability. This design provides a solid foundation for developing high-performance and durable flexible thermoelectric devices.

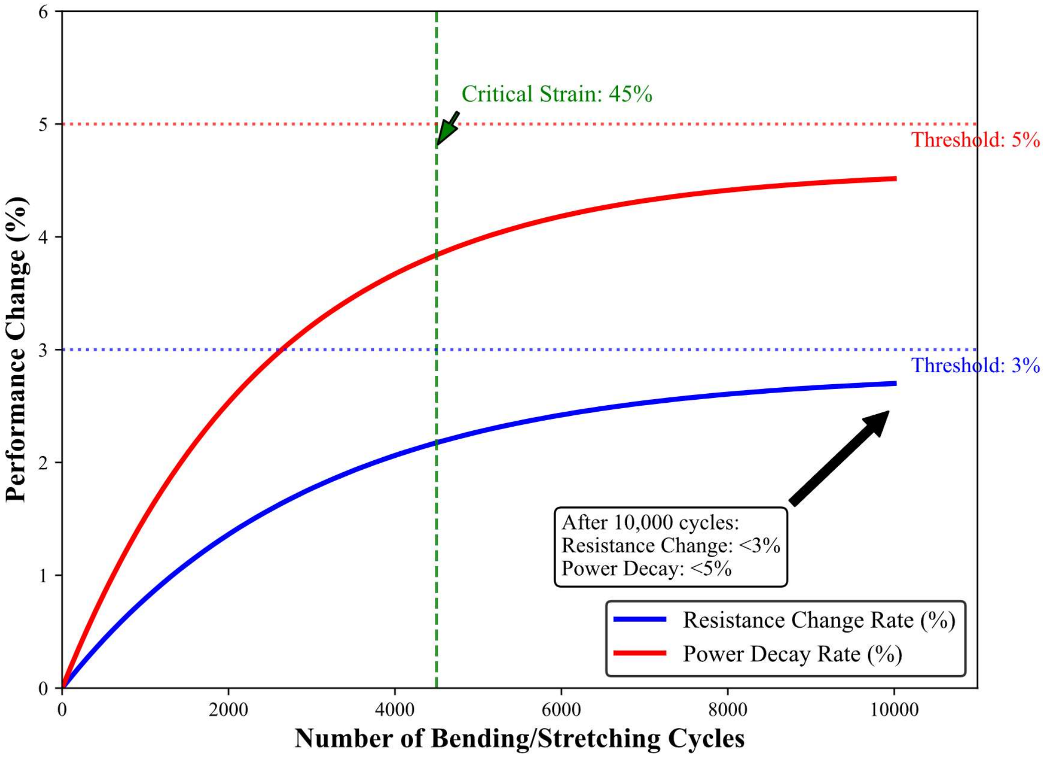

Figure 5. Variation trend of resistance change rate and power attenuation rate with cycle times

4.6. Bio-compatibility

In this study, the biocompatibility of the key materials of the device was evaluated, and the cytotoxicity (using L929 fibroblasts) and skin sensitization (guinea pig model) were tested according to ISO 10993 standard. The results showed that the cell survival rate of the material was over 90% (standard requirement > 70%), the skin irritation index was 0.2 (lower than the qualified limit of 0.8), and no erythema or edema reaction caused by sensitization was observed. All the test items meet the ISO 10993 standard, which shows that the device has good biocompatibility and is suitable for wearable application scenarios with long-term skin contact (see Table 3).

Table 3. Wearable application scenarios suitable for long-term skin contact

Test item	Result	Standard requirements
Cell survival rate	>90%	> 70% (non-toxic)
Skin irritation index	0.2 (no irritation)	≤0.8 (qualified)
Sensitization reaction	No erythema/edema	Nonresponse

5. Conclusion

(1) On the material level, the Bi_2Te_3/Sb_2Te_3 quantum dot superlattice thin film was constructed. By using quantum confinement effect, the Seebeck coefficient (258 μV/K) and carrier mobility (1250 S/cm) were significantly improved, while the thermal conductivity (0.9 W/mK) was reduced, and the comprehensive thermoelectric figure of merit (ZT) reached 1.32, which was nearly doubled compared with the traditional Bite thin film.

(2) On the structural level, a 3D spiral thermoelectric unit is designed, which significantly increases the effective thermoelectric arm length, reduces the interfacial thermal resistance (88 K·cm^2/W), and improves the thermoelectric conversion efficiency. At the system level, PCM and thermoelectric-photoelectric cooperative collection module are integrated, which effectively suppresses the heat flow fluctuation caused by human movement (temperature difference fluctuation < 1℃) and realizes the solar-thermal cooperative energy collection.

(3) The performance test results show that the power density of the optimized flexible thermoelectric device can reach 15 μW/cm^2 at a temperature difference of 5℃, which is two orders of magnitude higher than that of the traditional flexible device, and meets the ISO 10993 biocompatibility standard. In addition, the device also shows excellent flexibility/tensile reliability. After 10,000 cycles, the resistance change rate is less than 3%, the power attenuation rate is less than 5%, and the critical strain is 45%. These achievements provide a new technical path for developing high-performance and durable self-powered wearable devices, which is expected to promote the deep integration of the Internet of Things and AI technology and realize the intelligent and sustainable development of wearable devices.

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