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Thermoelectric Textile Materials

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http://dx.doi.org/10.5772/intechopen.75474

Abstract

Textile, as an intimate partner of human body, shows promising application in wearable thermoelectrics for body heat conversion. Compared with other widely studied flexible film thermoelectric materials, textiles having better air-permeability, wearability, and flexibility are more suitable for wearable electronics. In the past few years, many researches have focused on the design and fabrication of textile-based thermoelectric materials and generators. By integrating with high performance inorganic semiconductors and conductive polymers, fabrics or fibers will be given thermoelectric properties. Technologies of coating, printing, and even thermal drawing can be adopted in the fabrication of textile thermoelectric materials. With great design flexibility, various flexible textile generator structures can be obtained by using yarns or fabrics as thermoelectric legs, which will bring new inspirations for the future development of flexible thermoelectrics.

Keywords: thermoelectric, textile, flexible, wearable, fiber

1. Introduction

Recently, the rapid development of wearable electronic devices such as smart watches, wrist bands, smart glasses, and electronic skins has attracted extensive research interest of self-powered and wearable technologies [1, 2]. Energy harvesting technologies such as solar cell, piezoelectric, triboelectric, and thermoelectric technologies can be utilized to meet the requirement of self-powered system [3–7]. As one of the most promising energy harvesting strategies, thermoelectric technology has received extensive research attentions in recent years. In a thermoelectric system, heat especially waste heat, such as human body heat, geothermal energy, solar thermal energy, and the residual heat of motor engines, can be directly converted into electrical energy without any pollution [8]. A typical thermoelectric generator (TEG) is composed of several p-type and n-type thermoelectric materials connected parallelly



with conductors. When temperature difference exists between the two sides of thermoelectric materials, the charge carriers in the materials will be driven to flow from the hot side to the cold side, and thus a thermo-voltage can be formed. As key components in a thermoelectric system, the energy conversion efficiency of a thermoelectric material can be evaluated by a thermoelectric figure of merit, also called ZT value [9]. As shown in Eq. (1).

$$ZT = \frac{\sigma S^2}{\kappa} T \tag{1}$$

In this equation, σ is electrical conductivity, S is Seebeck coefficient, κ is thermal conductivity, T is absolute temperature, and ZT is a dimensionless value without upper limit. The value of $S^2\sigma$ is called power factor, which also can be used to represent the thermoelectric performance of materials where the impact of thermal conductivity is secondary. For example, in polymer-based thermoelectric materials, power factor is widely used, since most polymers have much lower thermal conductivity than inorganic semiconductors and then their thermoelectric performance will be mainly decided by the power factor.

Now, two main strategies can be adopted to improve the efficiency of thermoelectric system. One is to increase the ZT value of the thermoelectric materials, and the other is to adjust the structure of TEG for better utilization of heat energy. In past few years, the thermoelectric efficiency of bulk inorganic metal alloys has been improved significantly. A record-breaking high ZT of 2.6 achieved by SnSe single crystals at 923 K was reported in 2014, which gives thermoelectric generators a better application prospect in future [10]. Meanwhile, a new solar heat TEG system has been developed. By combining solar heat collection and thermoelectric conversion technologies, the new system could possess a peak efficiency of 7.4% [11].

Nowadays, with the rapid development of wearable and flexible electronics, the field of flexible thermoelectric materials and generators were growing dramatically. Various flexible thermoelectric materials were developed in form of films, papers, fabrics, and even fibers. Thermoelectric films are most studied among them, while the research about thermoelectric fabrics and fibers are just beginning very recently. Generally, these flexible thermoelectric materials are mostly made by organic polymers or polymer/inorganic semiconductor composites, so that they are easier to fabricate on large scales. However, the thermoelectric efficiency of these polymer-based flexible materials is much lower than the inorganic semiconductors. Hence, the methods to further improve the thermoelectric efficiency of these flexible materials will still be a hot research issue in future.

Although fabrics, fibers and polymer films are all flexible materials, textile thermoelectric materials have exhibited more competitive characteristics than simple films, especially on the wearable application. Textiles composed of different sets of fibers and yarns have excellent structure design flexibility, which allows them to meet the requirement of flexible, wearable, nontoxic, light weight, and even washable for wearable devices so that we can utilize our human body heat as power source. Until now, many pioneering research works have been conducted on this area and showed gratifying results. Therefore, in this chapter, the development of these textile thermoelectric materials and generators will be thoroughly described.

2. Textile thermoelectric materials

Textile is an ideal candidate to utilize our human body heat to generate power due to its excellent flexibility, wearability, comfort, and air-permeability properties, especially compared with those rigid bulk electronic materials and impermeable polymer-based flexible films. Thus, it is promising to develop textile material-based thermoelectric materials and generators to utilize our body heat. In the past few years, many research works have been done on both textile thermoelectric materials and fabric generators, which have paved the way and given us inspirations for future wearable thermoelectric systems.

2.1. Fabric-based thermoelectric materials

Fabric finishing and coating are mature technologies in textile industry that can be easily used for large-scale production. Thus, it is a cost-effective and practical way to added effective thermoelectric materials on fabrics for both organic and inorganic materials. In 2015, Yong Du et al. prepared a textile thermoelectric material by coating commercial polyester fabrics with PEDOT:PSS [12]. Their adopted a simple dip coating process to fabricate textiles with thermoelectric functions. **Figure 1** shows the dip coating process the prepared thermoelectric textile strip by Tong Lin. P-type PEDOT:PSS doped with DMSO was acted as effective thermoelectric materials. Commercial available polyester fabric was used as a flexible substrate. Both advantages of textiles and PEDOT:PSS were utilized. The prepared textile materials exhibit electrical conductivities ranged from 0.5–3 S/cm and Seebeck coefficient of 15.3–16.3 μ V/K at 300 K. The highest power factor of 0.045 μ Wm⁻¹ K⁻² was achieved at 390 K. Although the thermoelectric efficiency of this flexible textile thermoelectric material is not quite satisfied, this is

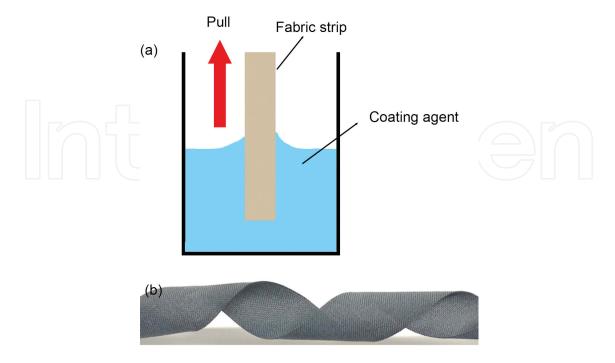


Figure 1. (a) A dip coating process and (b) PEDOT:PSS coated polyester fabric by Yong Du et al. [12].

still an initial attempt to applying fabrics into thermoelectric application. The low efficiency may also attribute to the used organic polymer thermoelectric materials. Choosing high efficient inorganic semiconductors especially their nano-sized counterparts as fabric coating materials, the thermoelectric efficiency coated fabrics could be further improved.

Another example is to use nano inorganic materials to produce thermoelectric fabric. In 2015, Chongjian Zhou et al. prepared a thermoelectric fabric made by inorganic nanowires and organic polymer composite [13]. A simple five-step vacuum filtration process was employed. $Cu_{1.75}$ Te nanowires sheet was fabricated by filtration, hot press, and annealing first, and then PVDF solution was drop cast on the nanowire sheet. After a heating process, a flexible composite material was formed. The composite structure and prepared fabric film are shown in **Figure 2**. The prepared flexible fabric film has a high electrical conductivity of 2490 S/cm, and a Seebeck coefficient of 9.6 μ V/K at room temperature, which resulted in a power factor of 23 μ Wm⁻¹ K². The performance of prepared fabric could keep steady after 300 cycles of bending tests. The results indicate that this easy to scale-up method is effective to fabricate flexible thermoelectric fabric and can be extended to other effective inorganic materials such as Bi₂Te₃, Ag₂Te, or Ag₂Se nanowires.

The preparation of fabric thermoelectric materials is simple and very easy to fabricate on large scale. The integration of inorganic high performance thermoelectric materials would give fabrics better thermoelectric performance than organic polymers. Although fabrics usually have the same 2D flat structure as polymer films, porous fabric structures would have better airpermeability and wearable comfort than polymer films, which is more appropriate for wearable devices.

2.2. Fiber/yarn-based thermoelectric materials

In addition to fabrics, fibers and/or yarns can be also fabricated into thermoelectric materials. Since fibers and yarns are the structural basis of fabrics, to offer fibers and yarns thermoelectric properties other than fabric itself will give more design flexibility for flexible textile TEGs. In 2012, Daxin Liang et al. reported a fiber thermoelectric material. PbTe nanocrystals were coated onto glass fibers via a dip coating process [14], as shown in **Figure 3**. The PbTe nanocrystal coated fibers have an electrical conductivity of 104.4 S/m, Seebeck Coefficient $1201.7 \, \mu \text{V·K}^{-1}$, and thermal conductivity of 0.228 W·m⁻¹·K⁻¹ at 300 K. The resulted power factor is 0.15 mW·m⁻¹·K⁻² at 300 K. It should be noted that the thin coating layer would lower the electrical conductivity of PbTe nanocrystals compared with PbTe bulk crystals.

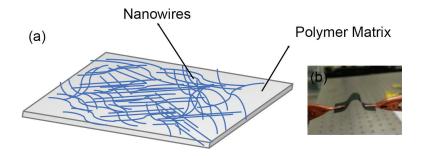


Figure 2. (a) The composite film structure and (b) the picture of flexible fabric film prepared by Chongjian Zhou et al. [13].

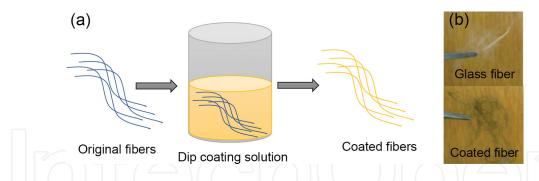


Figure 3. (a) Dip coating process of fibers and (b) comparison of uncoated and coated glass fibers made by Daxin Liang et al. [14].

In 2016, we proposed a water-processable thermoelectric coating material made of waterborne polyurethane, MWCNT, and PEDOT:PSS composite [15]. The optimal electrical conductivity and Seebeck coefficient could achieve ~13,826 S/m and ~10 μ V/K at room temperature respectively, and the resulted power factor is about ~1.41 μ Wm⁻¹ K⁻². Compared with other organic-solvent-based thermoelectric polymers, this water-based composite exhibits satisfactory thermoelectric performance and good processability. Then, we coated the prepared water-based composite on polyester and cotton yarns respectively, as shown in **Figure 4**. The results show that the fabricated thermoelectric composite can be successfully coated on textile yarns, and polyester filament is more suitable as coating substrate than staple cotton yarn. Besides, these coated yarns can be treated as thermoelectric legs and were further used to fabricate fabric TEG.

In 2016, Jae Ah Lee et al. using electro-spinning technology to fabricate thermoelectric nanofibers and then twisted into yarns [16]. Polyacrylonitrile (PAN) nanofiber sheets were electrospun on two parallel wire collectors. Bi₂Te₃ and Sb₂Te₃ were selected as n-type and p-type thermoelectric materials, and deposited on two sides of the PAN sheets. After a twisting process, thermoelectric yarns can be formed, as shown in **Figure 5**. The highly porous structure of the electrospun yarns are quite resistant to mechanical damage, which enables the yarns to be knitted and woven into fabrics without the significant changes in their electrical conductivity and thermopowers.

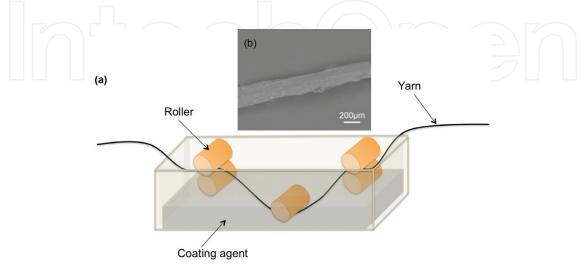


Figure 4. (a) Yarn coating process and (b) polyester yarn coated with waterborne polyurethane thermoelectric composites made by Jinlian Hu [15].

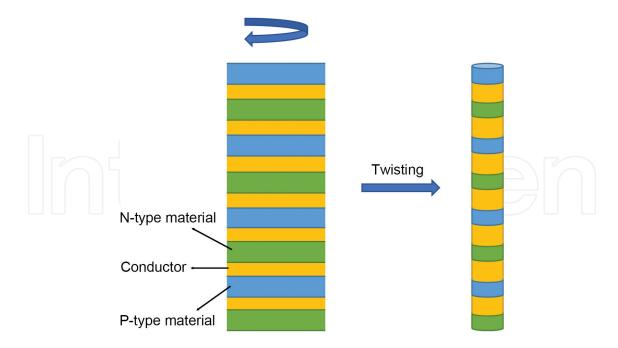


Figure 5. Schematic illustration of the conversion of thermoelectric sheet into a yarn.

The studies of yarn and fiber thermoelectric materials are just beginning. The same coating process of fabrics can be further used to prepare fiber and yarn thermoelectric materials. Additionally, more complicated fabrication technique such as electrospinning and twisting process can also be adopted to prepare fiber and yarn materials with better performance. It can be imagined that there will be more ingenious methods in future to fabricate high performance thermoelectric fiber or yarn materials, so that a fully textile-based thermoelectric generator can be achieved.

3. Textile thermoelectric generators

Generally, a traditional inorganic TEG possesses a sandwich structure. Several n-type and p-type thermoelectric bulk materials are alternatively arranged and connected in series with conductors in the middle part of TEG. Ceramics are used at the top and bottom layers to avoid short-circuit between the metal interconnects. This sandwich structure helps to maintain an excellent heat exchange with surroundings. Temperature difference will be generated along the thickness direction of sandwich TEG, which is necessary for wearable thermoelectrics to some extent. However, it is difficult to apply this sandwich structure in flexible film materials. The structure of a typical organic film TEG is a two-dimensional plane. Although p-type and n-type film thermoelectric legs are still alternatively connected in series by flexible conductors, the temperature difference in this structure will only exists along the film length direction rather than along the thickness direction, since the dimension limitation in film thickness direction cannot sustain the temperature difference. Thus, the flexible 2D film generator will be challenged in wearable application. Textiles TEG with various structure design flexibility may provide one solution for this problem.

3.1. Two-dimensional textile TEGs

A 2D flat TEG structure is also adopted in the first generation of textile TEGs. In 2012, C. A. Hewitt et al. developed a PVDF/CNT composite-based fabric TEG. Several n-type and p-type CNT composite films were alternatively arranged between the insulated PVDF films. The PVDF films are shorter than the CNT composite films. Thus, n-type and p-type CNT composites could form p-n junctions to connect the generator legs by hot press the stacked films, and resemble a felt fabric TEG [17]. The schematic structure of they prepared fabric TEG is shown in **Figure 6**. The prepared TEG composed of 72 layers fabric could generate power about 137 nW with an internal resistance of 1270 Ω .

In 2015, Yong Du et al. prepared a 2D fabric TEG by using PEDOT:PSS coated polyester fabric strips [12]. To fabricate the TEG, a whole PEDOT:PSS coated polyester fabric was cut into several strips first. Then, these strips were further adhered on a polyester fabric substrate by silver paint and connected in series with silver wires. Thus, a fabric TEG only composed of p-type materials can be prepared. The TEG arrangement is shown in **Figure 7**. Temperature difference will generate along the fabric length direction. The fabric TEG prototype consisting of five fabric strips could generate 4.3 mV when temperature difference ΔT is 75.2 K. The maximum output power P_{max} could achieve 12.29 nW.

The same arrangement can also be achieved by thermoelectric yarns. In 2017, Ryan et al. reported a highly durable thermoelectric silk yarn made by dyeing with PEDOT:PSS, which could be produced with a length of up to 40 m and keep steady after repeated machine washing and drying [18]. Then, they embroidered these yarns into a felted wool fabric substrate and connected them end-to-end with silver wires to form a fabric TEG. The structure is illustrated in **Figure 8**. In an in-plane fabric TEG prototype composed of 26 yarn legs, the internal resistance is about 8.7 K Ω , and the output voltage of $V_{out}/\Delta T$ is about 313 μV K⁻¹ when temperature difference ΔT is about 120°C. An output current of 1.25 μA can be observed when ΔT is 66°C, and resulted a maximum power output P_{max} of ~12 nW.

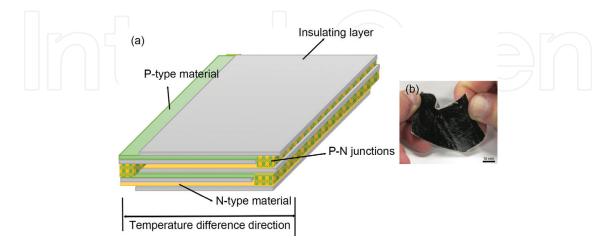


Figure 6. (a) Alternative arranged multilayer fabric TEG structure and (b) fabric film TEG prepared by Hewitt et al. [17].

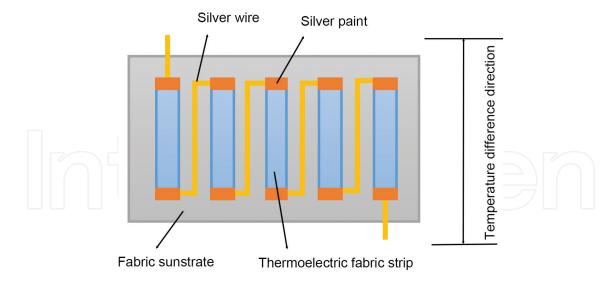


Figure 7. Arrangement of 2D fabric TEG composed of only p-type materials.

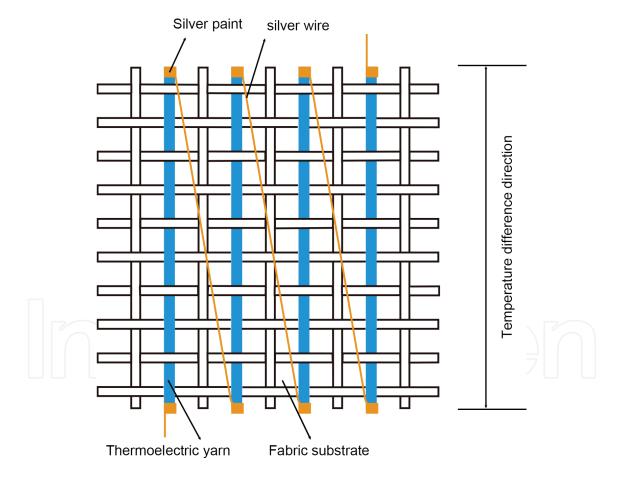


Figure 8. Thermoelectric yarn arrangement in a fabric substrate.

In general, 2D fabric TEG is a first attempt to apply textile structures to the design of flexible TEGs. Although these initial 2D fabric TEGs have almost the same in-plane structure with film TEGs, fabric TEGs can be easily rolled up, bent, twisted, and are permeable to air and moisture, making them more flexible and comfort to wear.

3.2. Three-dimensional textile TEGs

In addition to the initial 2D textile TEG structures, researchers also developed several 3D textile TEGs similar to the classical sandwich bulk TEG. In 2014, Kim et al. fabricated a wearable TEG on a glass fabric by screen-printing technology [19]. The device structure and is shown in Figure 9. The inorganic Bi, Te, and Sb, Te, thermoelectric materials were printed on a glass fabric substrate first. Then, the fabric containing an array of eight thermocouples was connected by several cooper foils, and finally encapsulated with PDMS. The fabricated TEG exhibits a high output power density of 3.8 mWcm⁻² and 28 mWg⁻¹ at $\Delta T = 50$ K. Besides, the TEG could endure repeated bending for 120 cycles.

Another research reported a silk fabric-based TEG similar to the Kim's structure in 2016. Zhisong Lu et al. deposited nanostructured Bi₂Te₃ and Sb₂Te₃ synthesized by hydrothermal method on two sides of a commercial available silk fabric [20]. The deposited p-type and n-type thermoelectric materials were further connected with silver foils to form a flexible TEG using a similar arrangement of Kim et al. [19]. The prototype containing 12 thermocouples could generate a maximum thermos-voltage of ~10 mV and output power of ~15 nw under a temperature difference of 35 K. The power output performance can sustain stable even during 100 cycles of bending and twisting.

Jae Ah Lee et al. using textile structure designed a new type of fabric TEG, which can utilize thermal energy along fabric thickness direction. Both knitting and weaving technology can be employed to fabricate this fabric TEG [16]. Several p-type and n-type yarns prepared by electro-spinning were arranged in the fabric according to the predesigned patterns. In knitted structure, p-type and n-type yarns were alternatively arranged and connected in series. In a plain weave structure, several single yarns containing metallic connected n-type and p-type components were woven into fabrics with insulating yarns. In addition, these single yarns should be carefully placed in correct way to ensure the right contact between p-n junctions and hot/cold surfaces. Figure 10 illustrates the fabric TEG in knitted and woven structures respectively. The best output power of the prepared fabric TEG could achieve 8.56 Wm⁻² at a temperature difference of 200°C. This study is the first attempt to utilize fabric structures to realize textile TEG that suitable for fabric thickness power generation.

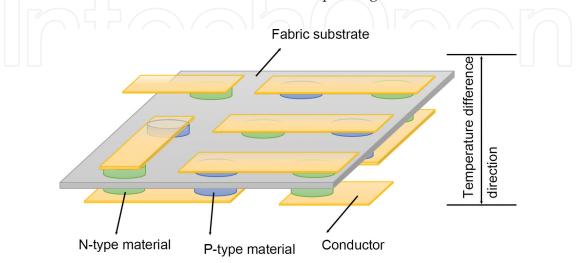


Figure 9. Arrangement of fabric TEG that allowing generate temperature difference along fabric thickness direction.

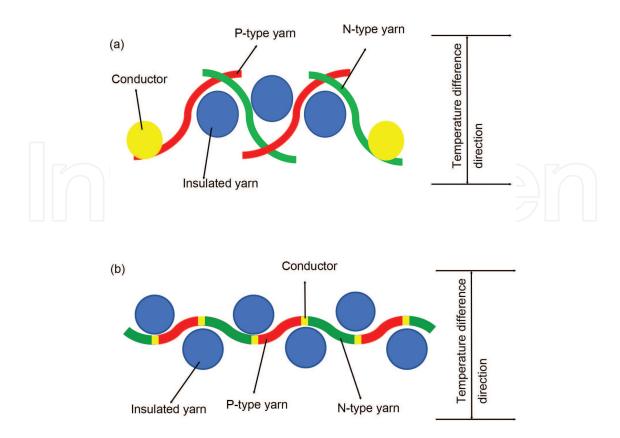


Figure 10. (a) Thermoelectric yarn arrangement in knitted fabric structure and (b) thermoelectric yarn arrangement in woven fabric.

In the following 2017, we designed a new flexible 3D fabric TEG which have a sandwich structure, so that the temperature difference could be generated along fabric thickness direction [21]. A 3D spacer fabric is composed of two separate fabric layers on the top and bottom of the fabric, and a set of pile yarns in the middle used to connected the top and bottom fabric layers. Compared with 2D materials, the sandwich 3D structure has more dimensions in thickness direction, so that the temperature difference can be sustained along the fabric thickness direction, and the flexibility of the TEG can be retained due to its fabric nature. Therefore, 3D fabric would be a good candidate as substrate for flexible and wearable TEG. **Figures 3–5** show the designed 3D fabric TEG. A prototype was fabricated by embroidered p-type and n-type coated polyester yarns into a 3D spacer fabric. The 3D fabric TEG consisting of 10 couples of thermoelectric yarns would generate a thermo-voltage of ~800 μ V and output power of ~2.6 nW at a temperature difference of 66 K.

3.3. Other textile TEGs

There are also some new creative textile TEGs developed very recently. In 2017, Ting Zhang et al. demonstrated a novel crystalline ultralong thermoelectric fiber and two TEG prototypes [22]. Thermal drawing technology was employed to integrate high performance thermoelectric materials in a fiber-like carrier, which is also a physical thermal size-reduction strategy. P-type Bi_{0.5}Sb_{1.5}Te₃ and n-type Bi₂Se₃ nanowires were used as core thermoelectric materials, and borosilicate glass tube with a T_g slightly higher than the thermoelectric nanowires were

used as cladding materials to form a core-shell structure fiber. The fabricated thermoelectric fibers are long, flexible, intrinsically crystalline, and mechanically stable, which can be further applied in flexible TEGs. The two pairs of p-n fibers composed TEG has an internal resistance of 410 Ω, and could generate an open-circuit voltage of 24.2 mV, a short-circuit current of 59.1 μA, and a maximum output power of 0.36 μW, when applying a temperature difference of 50 K. To demonstrate the advantages of fiber's length and flexibility, a thermoelectric cup and a thermoelectric pipe were fabricated by wrapping several pairs of p-n thermoelectric fibers on the curved surface.

In 2016, Abu Raihan Mohammad Siddique et al. employed a manual dispenser printing technique to fabricate a flexible fabric TEG [23]. (0.98Bi,0.02Sb),(0.9Te,0.1Se), was used as n-type material, (0.25Bi,0.75Sb)₂(0.95Te,0.05Se)₃ was used as p-type material. Two TEG prototypes containing 12 pairs of p-n thermoelectric materials were fabricated by printing the selected thermoelectric materials on polyester fabric. The fabricated prototypes consisting 12 pairs of n-type and p-type legs and connected in series with silver wires. The best open circuit voltage and output power were 23.9 mV and 3.107 nW, respectively, under a temperature difference of 22.5°C. The wearing test on human body proves that the fabricated prototypes are very flexible, twistable, and durable with the substrate as well as conforming well to the human body.

In 2017, Jaeyoo Choi et al. reported a flexible and ultralight TEG-based on carbon nanotube yarn (CNTY) with excellent thermoelectric performance [24]. A high electrical conductivity of 3147 S/cm was achieved by a highly aligned CNTY, which has increased longitudinal carrier mobility. In their work, the CNTY exhibits multifunction in the TEG. N- and p-types dopants of polyethylenimine and FeCl, were alternatively doped into CNTY to generate n- and p-type legs in the CNTY device respectively. Between the doped regions, highly conductive undoped CNTY regions were acted as electrodes to connect and minimize the circuit resistance. Thus an all-carbon TEG without additional metal deposition process can be formed. A prepared TEG prototype containing 60 pairs of n- and p-type doped CNTY exhibits the maximum power density of 10.85 and 697 μ W/g at Δ T of 5 and 40 K, respectively.

Screen printing technology is a low-cost fabrication technique that is applicable to large scale production. Zhuo Cao et al. demonstrated that screen printing technology can be used to fabricate BiTe/SbTe-based TEGs for room temperature energy harvesting applications on flexible substrate [25]. Although the fabricated TEG contained only eight pairs of thermoelectric legs, the screen printing process and materials flexibility allow longer strips with larger numbers of thermocouples to be connected in series and rolled up to form a coil TEG and enabling the power output to be increased. Thus, it is also a practical fabrication method to prepare fabricbased TEGs on large scale.

Besides the thermoelectric performance of thermoelectric materials, temperature difference is also an important impact factor on the thermoelectric efficiency of TEGs. A higher △T would increase the output power of TEGs. However, the temperature difference between human body and environment are usually in the range of 1.5–4.1°C, which is unfavorable for body heat utilization. It is necessary to find ways to enlarge the temperature difference for wearable TEGs. In 2017, Yeon Soo Jung et al. proposed a novel approach to increase the $\triangle T$ of conventional wearable TEGs, they present a wearable solar TEG possessing a high $\triangle T$ value of ~20.9°C by introducing a local solar absorber and thermoelectric legs on a polyimide substrate [26]. The prepared solar absorber is composed of a five-period Ti/MgF, superlattice. The structure and thickness of each layer was carefully designed to absorb sunlight at maximum extent. A dispenser printing technique was employed to prepare thermoelectric legs on the substrate. The n- and p-type ink were made by alloyed BiTe-based powders and Sb₂Te₃based sintering additive dispersed in glycerol. A wearable TEG prototype consisting of 10 pairs of thermoelectric legs exhibits an open circuit voltage of 55.15 mV and an output power of 4.44 μ W when exposed to sunlight. The generated high Δ T of ~20.9°C between the hot solar absorber and cold edges is also the highest $\triangle T$ value of all wearable TEGs reported to date. In 2016, Melissa Hyland et al. also reported a wearable TEG device with optimized heat spreaders to increase the $\triangle T$ of wearable TEG [27]. The integration of heat spreader would improve the dissipation of heat and cooling throughout the wearable TEG. In their design, a three-layered device structure were used. Two flat heat spreaders were equipped on the top and bottom of TEG respectively. The sandwich structure was chosen as the final design due to its high efficiency and low form factor.

4. Conclusion

With the rapid growth in wearable and flexible electronics, the demand in flexible self-power technologies are also increased. Thermoelectric energy conversion technology shows great potential in make use of our human body heat to generate power, which would be an ideal power source candidate for wearable electronic systems. In most of the developed flexible thermoelectric materials and generators, textile-based thermoelectric materials and TEG have unique advantage in body heat energy conversion due to their excellent air permeability, flexibility, and wearing comfort, especially for natural materials such as silk and cotton, which shows good biocompatibility. Therefore, a fabric-based thermoelectric generator with great wearability would overcome the wearable difficulty of existing organic film generators, which shows promising application in flexible and wearable self-powered electronic systems by harvesting body heat to generate electricity. In the past years of study, many textile thermoelectric materials were prepared in forms of fibers and fabrics including some traditional inorganic semiconductors and the newly developed organic polymers and composites. Compared with 2D fabrics, fiber-based thermoelectric legs would give more design flexibility for the flexible TEGs. Besides, some pioneering researches also designed and fabricated several novel textile TEGs with excellent flexibility and thermoelectric performance. Various TEG structures such as 2D generator making use of in-plane temperature difference and 3D generators generating temperature difference along fabric thickness directions are included. These textile-based TEGs with multiple structures are more practical and suitable for wearing than the previous widely studied film TEG, which shows promising application in future self-powered wearable system driven by body heat. The study of flexible and wearable thermoelectric materials and generators is just beginning. The efficiency of textile thermoelectric materials and generator need to be further improved for real application. These creative works bring many inspirations for the future explorations in thermoelectric field.

Acknowledgements

This work was supported by University Research Grants Council, PolyU 5162/12E, Shen Zhen Government Key Incubation Fund, JC201104210132A, National Key Technology R & D Program Project, The Ministry of Science and Technology of P.R.C, 2012BAI17B06, and National Natural Science Foundation of China, 51673162.

Conflict of interest

There are no conflicts of interest to declare.

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