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A novel design of wearable thermoelectric generator based on 3D fabric structure

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Abstract

A flexible and wearable thermoelectric generator could enable converting our human body heat into electrical power, which help to realize a self-powered wearable electronic system. To overcome the wearable difficulty of existing flexible film thermoelectric generators, a novel 3D fabric thermoelectric generator structure are designed in this study. By using a 3D fabric as substrate and yarns coated with thermoelectric materials as legs, a wearable and flexible thermoelectric generator can be realized. The designed generator has a sandwich structure similar to the classical inorganic generator, which allows generating temperature difference in the fabric thickness direction, thus enable people to wear and show promising application in body heat conversion. Then, to verify the effectiveness of designed generator structure, a prototype was fabricated and by using a lock-nit spacer fabric as substrate and yarns coted with waterborne polyurethane/carbon nanotube thermoelectric composites as legs. The results suggested that the fabricated spacer fabric TEG prototype could work successfully, although the performance of this prototype is in a low level. To further improve the efficiency of 3D fabric generator and apply it in wearable electronics in future, highly efficient inorganic thermoelectric materials can be applied, and modifications on the conductive connections can be made.

Keywords: thermoelectric generator, 3D fabric, spacer fabric, wearable electronics

1. Introduction

Thermoelectric generators (TEGs) that are capable of direct converting temperature differences into electrical voltages have attracted extensive research interests due to their mechanical stability and pollution-free characteristics.[1] For example, during the past few decades, large-scale TEGs have been successfully used as reliable power sources for deep space probes of NASA.[2] In addition to such large-scale devices, in recent years, this green technology shows promising applications in small-scale wearable, self-powered electronics that can convert body heat into electrical power.[3] Unlike the traditional rigid TEGs made of bulk inorganic thermoelectric materials for large scale power conversion, the TEGs used for small scale electronics are usually flexible and made of organic materials. Therefore, in the past few years, various organic materials such as conjugated polymers and conductive polymer composites have been developed for thermoelectric application.[4–8] However, until now, the efficiency of organic thermoelectric material is still much lower than the inorganic ones, which restricts the efficiency of flexible TEG made of organic materials.

Compared with the development of flexible thermoelectric materials, the study of flexible and wearable TEGs is still in its infancy. Now, unlike the traditional inorganic bulk material based TEG having a 3D sandwich structure, most of the polymer based flexible TEGs use a 2D thin film structure due to the very low thickness of polymer materials. [9–12] In this structure, p-type and n-type thermoelectric thin films are alternatively arranged and connected electrically in series in a plane. For example, Hewitt et al. reported a 2D flexible film TEG by using alternatively arranged p-type and n-type PVDF/CNT composite films as thermoelectric materials.[13] Apart from thin film polymers, fabric with superior permeability, flexibility and wearablility is ideal place to utilize body heat. Some studies have reported fabric TEGs. Du et al. fabricated a fabric TEG by series connecting several PEDOT:PSS coated fabrics.[14] Similar to those polymer thin films, this fabric TEG also has a 2D flat structure that harvesting in-plane direction thermal energy. Recently, some pioneering work demonstrated new flexible TEGs by using highly efficient inorganic thermoelectric material that could harvest thermal energy along desired thickness direction, but a high temperature condition and complicated fabrication process were needed in these study. Kim et al. reported a flexible TEG by screen-printing highly efficient inorganic Bi_2Te_3 and Sb_2Te_3 thermoelectric materials on glass fabric and encapsulated with rubber sheet. [7] Since this process needs a fabrication temperature higher than 500°C, only inorganic glass fabric can be used which is not the fabric for garment. Lee et al. fabricated woven and knitted thermoelectric textiles by using electrospun PAN with alternatively deposited Bi_2Te_3, Au and Sb_2Te_3 as yarns, whereas the fabrication process is quite complicated due to the complex fabric structure design. [15] To the best of our knowledge, almost all the previous study about organic material based fabric TEG is using 2D flat structure and harvesting thermal energy along in-plane direction.

Moreover, the mostly used 2D flat structure would cause wearable difficulties, which restricts their application in wearable field. In wearable situation, a temperature difference exists between the human body and the environment, thus the heat flux direction is perpendicular to our skin. However, 2D flat TEG using the length direction temperature gradient will not work when people wearing it, as the length direction of the film is parallel to our skin and temperature difference will not be generated. In this case, although an organic film TEG is flexible, it is difficult to apply it into wearable situation. It is therefore necessary to develop an easy-to-fabricate new structure in which both flexibility and wearability can be achieved. Herein, we describe a new 3D fabric based flexible and wearable TEG structure that allows generating temperature difference along fabric thickness direction, like in classical inorganic TEG. 3D fabric TEG was fabricated by alternatively embroidering p- and n-type organic thermoelectric composites coated yarns into a spacer fabric substrate and connected electrically in series via conductive paint. This easy-to-fabricate design could solve the wearable difficulty of film TEG, and is promising to be applied in body heat conversion application.

2. Experiment

2.1 Materials

Nonionic waterborne polyurethane (NWPU) is the same as it was synthesized in our previous study. [16] MWCNT was purchased from SouthWest NanoTechnologies, Inc. (CoMoCAT®, with purity>95%, O.D. × L: 6-9 nm × 5 μm). N-doped MWCNT was purchased from Times Nano Co. (purity 95wt%, N content: 2.98wt%, O.D. × L: 30-50 nm × 10~30μm). PEDOT:PSS (CLEVIOS™ PH1000, solid content 1.3%) were purchased from Heraeus Clevios GmbH. DMSO was purchased from Sigma Aldrich. Polyester yarn and spacer fabric were purchased from a local retailer. Silver paint (SCP) was purchased from Electrolube Co.

2.2 Thermoelectric composites coating on polyester yarns

To fabricate thermoelectric yarns, nonionic waterborne polyurethane (NWPU) based multiwalled carbon nanotube (MWCNT) thermoelectric composites were used to coat on commercial polyester yarns. The thermoelectric properties of NWPU thermoelectric composite films and yarn coating performance have been reported in our previous study.[16] In this study, NWPU/PEDOT:PSS/multi-walled carbon nanotube (MWCNT) composite containing 20wt% MWCNTs and 1:4 ratio of MWCNTs to PH1000 (doped with 5wt% DMSO) was used for the p-type coating agent, and NWPU/nitrogen doped multi-walled carbon nanotube (N-MWCNT) composite containing 30wt% of N-MWCNTs was used for the n-type coating agent. These composites were directly coated on commercial polyester yarn for five times via a dip coating method.

To fabricate these composites, MWCNTs/(N-MWCNT) were first dispersed into the PEDOT:PSS dispersions (containing 5wt% DMSO)/(water) respectively and sonicated for 30 min, then NWPU was added and stirred at 60°C for 2 hours, and finally suffered another sonication for 20 min. The polyester yarns were dipped into the fabricated p-type and n-type composite dispersions respectively, and dried in an oven at 100°C. The coating procedure was repeated for five times.

2.3 Fabrication of 3D fabric TEG

Figure 1 illustrates the fabrication process of a 3D fabric TEG. A spacer fabric substrate and coated p-type and n-type yarns are raw materials that need to be prepared in advance. In the first step, the prepared p-type and n-type yarns were embroidered into the spacer fabric substrate and arranged alternatively. In the second step, silver paint was patterned on top and bottom of the loops of yarns to connect every adjacent p-type and n-type yarns in series. In the final step, two copper wires were attached on the first p-type and last n-type yarns as output pins of the fabric TEG. Besides, silver paint was applied to the connections between yarns and copper wires to reduce contact resistance. Then, a 3D fabric TEG can be obtained.

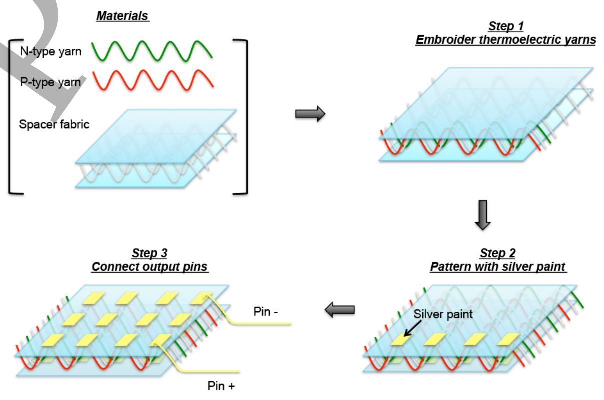

Figure 1 Fabrication process of 3D fabric TEG

2.4 Characterizations

Surface morphologies of coated yarns were observed by FE-SEM (JEOL, Model JSM-6335F). The spacer fabric thickness was measured with a vernier caliper. To generate a temperature difference across the 3D fabric TEG and investigate its output characteristics, a heater was placed under the bottom of fabric TEG and top surface of the TEG was exposed to air. Temperatures of the top and bottom side of TEG were simultaneously measured by two K-type thermocouples. As the temperature difference was increased from 4.5K to 66K, the output characteristics were measured by matching the external load with the internal resistance of the fabric TEG by using a Keithley 2000 multimeter (Keithley Instruments Inc., Cleveland, USA). The demonstration of body heat conversion was performed via attaching the fabric TEG on a human wrist with insulated double-side adhesive tape and measuring the generated thermal voltage. An ice water bottle was used to cool the fabric layer exposed to air to create temperature difference.

3. Results and discussion

3.1 3D Fabric TEG design

By introducing 3D fabric structure, the TEG designed in this study has a sandwich structure that allows generating temperature difference along thickness direction. Figure 2 illustrates the structure design of 3D fabric TEG in three views. In the 3D view, the blue 3D grid represents a 3D fabric, the green and red lines represent the p-type and n-type yarn legs respectively, and the yellow areas are the conductive paint. In this design, 3D fabric provides a sandwich substrate; p-type and n-type thermoelectric yarns are arranged alternatively in parallel in it. Electro-conductive paints are incorporated on top and bottom side of adjacent p-type and n-type yarn loops to connect these yarn TE-legs in series to form a complete TEG. With the support of 3D fabric substrate, a sandwich TEG structure like classical inorganic TEG can be realized and temperature difference could exist between the top and bottom layers of 3D fabric. From the front view, it can be observed that the p-type and n-type yarns are alternatively arranged and connected similar to that in 2D film TEGs. In this structure, every single yarn corresponds to one leg in TEG. When a temperature difference is generated between the top and bottom layers of the 3D fabric, the charge carriers in the embroidered p-type and n-type yarns will be driven to defuse from the hot side to the cold side, and generate a thermal voltage. From the side view, the yarn morphology is revealed in the out of plane direction, which represents the 3D flexible structure of TEG.

It can be found that the designed fabric TEG has simple structure. To fabricate such TEG, an appropriate 3D fabric substrate and yarn legs are needed. In comparison with conventional 2D fabric, 3D fabric exhibits a typical sandwich structure, and full three-dimensional reinforcement can be provided by a single fabric.[17,18] A 3D fabric consists of two separated fabric layers that are connected with a set of pile yarns in the fabric thickness direction, which can be subjected to out-of-plane loadings due to the extra length provided by the pile yarns in the thickness dimension. Such 3D fabrics with good compressibility, thermoregulation capability, and wide structure variations are widely used in the design of protective clothing, medical textiles, sports wear, and many complex composites.[19,20] However, those 3D fabrics having large mesh surface structures exhibit high air and water vapor permeability that cannot sustain the temperature difference between the top and bottom sides and thus are inappropriate as substrate in this fabric TEG. Therefore, those 3D fabrics having a tight surface structure are favored, since they can provide a good thermal insulation to the fabric TEG, which is necessary to sustain a good working condition for the TEG. [21–25] Another essential component for the 3D fabric TEG is the thermoelectric yarn leg. Theoretically, these yarn legs can be produced by any functional yarn fabrication methods such as spinning or coating, depending on the processing characteristics of selected thermoelectric materials. In this study, the yarn legs were fabricated by a simple coating method. NWPU with a great viscosity can be easily adhered on yarn surface and has been widely used in textile functional coating industry, thus it is one of the most simple way to give thermoelectric properties on yarns by direct coating NWPU based flexible thermoelectric composites on yarns. [26–30]

In general, similar to that in inorganic sandwich TEGs, the temperature difference could exist along the TEG thickness direction in this 3D fabric TEG design, and furthermore the flexibility of the TEG can be retained due to the flexible nature of fabric substrate. Besides, The designed 3D fabric TEG can be easily fabricated due to its simple device structure, which is beneficial to large-scale production.

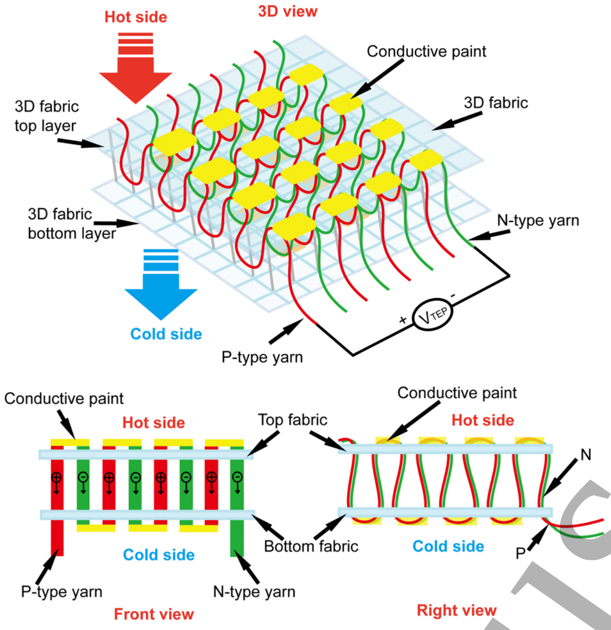

Figure 2. Structure design of 3D fabric TEG;

3.2 Spacer fabric TEG prototype Morphology

To assess the effectiveness of designed 3D fabric TEG structure, a spacer fabric TEG prototype comprising 10 couples of p-type and n-type yarn legs with size of 6 cm × 6 cm × 7 mm was fabricated. The morphologies of the raw materials and spacer fabric TEG prototype are presented in Figure 3. In this prototype, a warp knitted spacer fabric with lock-nit surface structure was used as the 3D fabric substrate, as shown in Figure 3 (a). Two separate lock-nit fabrics were incorporated as top and bottom layers and were linked together by the middle spacer monofilaments. Tightly knitted top and bottom layers can maintain a good thermal insulation in the middle space of the spacer fabric, which is beneficial in keeping a temperature difference between the top and bottom layers of spacer fabric. Figure 3 (b) and (c) present the SEM morphologies of p-type and n-type coated yarns respectively. The p-type composite was evenly covered the entire surface of polyester yarn, which proves a good coating layer was formed. However, the n-type composite was only partly covered the yarn, which would reduce the efficiency of coated yarn. This may attribute to the high CNT contents and lack of PEDOT:PSS that decrease the film forming ability of the composite. Yet, until now, the study of organic n-type thermoelectric material is still in initial stage, which restricts the options of n-type material and also limits the efficiency of organic TEG.

Figure 3 (d)~(e) show the morphologies of spacer fabric TEG prototype under two different processing stages. In Figure 3 (d), ten pairs of coated p-type and n-type yarns were embroidered alternately in the lock-nit spacer fabric substrate. In Figure 3 (e), conductive silver paint was patterned on the top and bottom yarn leg loops to connect the adjacent two p-type and n-type yarns. Two copper wires were attached onto the first p-type and the last n-type yarn legs as the external connect pins. Figure 3 (f) exhibits the flexibility of prepared fabric TEG prototype. It can be observed that the fabric TEG can be easily bent.

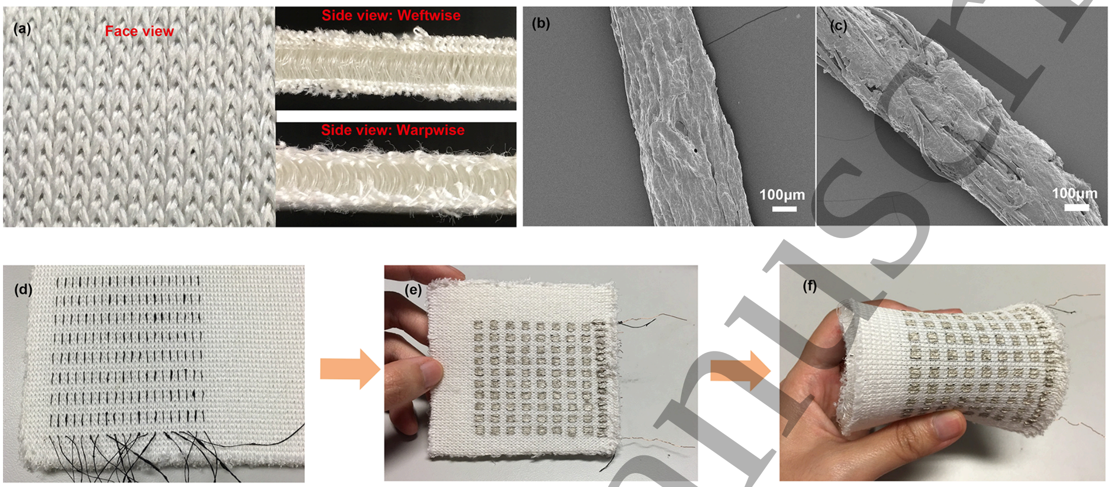

Figure 3. (a) Morphology of lock-nit spacer fabric; (b) P-type coated yarn; (c) N-type coated yarn; (d) Yarn legs embroidered into the spacer fabric matrix; (e) Silver paint was patterned to connect yarn legs; (f) The flexibility of fabric TEG prototype

3.3 Thermoelectric performance of the prototype

The output characteristics of the spacer fabric TEG prototype are demonstrated in Figure 4. Figure 4 (a) and (b) present the output voltage and power of the fabric TEGs with different numbers of yarn couple modules under different temperature gradients. It can be observed that both the thermo-voltage and output power of the spacer fabric TEG increased with the increase of ΔT, which indicates the fabricated prototype is effective in thermoelectric conversion and proves the designed 3D fabric TEG structure is feasible. Besides, the thermo-voltage and output power of the prototype also increased when incorporating more yarn couples in the fabric. However, the fabricated spacer fabric TEG prototype comprising 10 couples of coated yarns only generate an open circuit voltage of ~800 μV and output power of ~2.6 nW at ΔT= 66 K. This is primarily due to the small amount of coating layers on limited yarn surface areas, which cause the content of thermoelectric materials on yarns is much lower than that in film sample. Increasing the thickness of coating layers or using thicker yarn substrate would increase the content of thermoelectric materials on yarns, and it is possible to enhance the output performance of fabric TEG. But generally, the efficiency of organic thermoelectric materials is much lower than that of inorganic ones. This also means that applying inorganic thermoelectric materials to fabricate yarn legs would significantly improve the performance of fabric TEG, as those high performance fabric TEGs usually use inorganic thermoelectric materials. [7,15] Since the polyester yarn used in this study is a commercial thinner yarn, another reason for the poor performance may be the small contact areas between adjacent p-type and n-type yarns that would cause the increase of electrical and thermal contact resistance in the fabric TEG. To reduce the impact of contact resistance, future amendment should increase the contact areas between yarn legs. Incorporating yarn legs with bigger fineness could increase the yarn surface areas, which is benefit to increase the contact areas between yarns and reduce the contact resistance in the TEG. Meanwhile, new surface structures of spacer fabric should be applied to fit for the embroidering of thicker yarns.

Figure 3 (c) and (d) show the internal resistance and thermo-voltage of fabric TEG with increasing couples of yarn modules at 66 K. Both the internal resistance and thermo-voltage of the fabric TEG increased linearly with the increasing number of yarn modules. When the generator has 10 couples of yarn modules, the internal resistance is ~240 Ω. Since the conductive connections in the fabric TEG may be affected in real wearable situation, we also investigate the internal resistance stability of the fabric TEG prototype under bending stress along two different directions, weftwise and warpwise respectively, as shown in Figure 3 (e). It can be observed that the internal resistance of fabric TEG prototype is slightly increased with the increase of bending radius on both directions, but the change is less than 10%. The increment of internal resistance in weftwise direction is a little higher than that in warpwise direction. As silver paint lack of flexibility, cracks would appear during the bending process, which would damage the quality of conductive joint connection in the TEG, and thus cause the increase of internal resistance. To solve this problem, the silver paint can be patterned under fabric pre-bending state not the original flat state, which could ensure the completeness of silver paint under bending or tensile state, and improve the stability of TEG in wearable situation. Similar pre-stretching method has been used in many stretchable electronics.[31,32] Although the fabric TEG containing 10 couples of yarn legs generates only a relatively small amount of power, further to increase the fabric TEG areas will enhance linearly the performance of this generator. Large array of yarn legs can be easily embroidered into this fabric structure. According to the relationship of thermo-voltage per area of the fabric TEG shown in Figure.3 (f), it can be predicted that an adult man (with height of 180 cm, weight of 75 kg, and body surface area of 2.0s m^2) wearing the fabric TEG from head to foot would generate a thermo-voltage of 540 mV under a temperature difference of 66K.

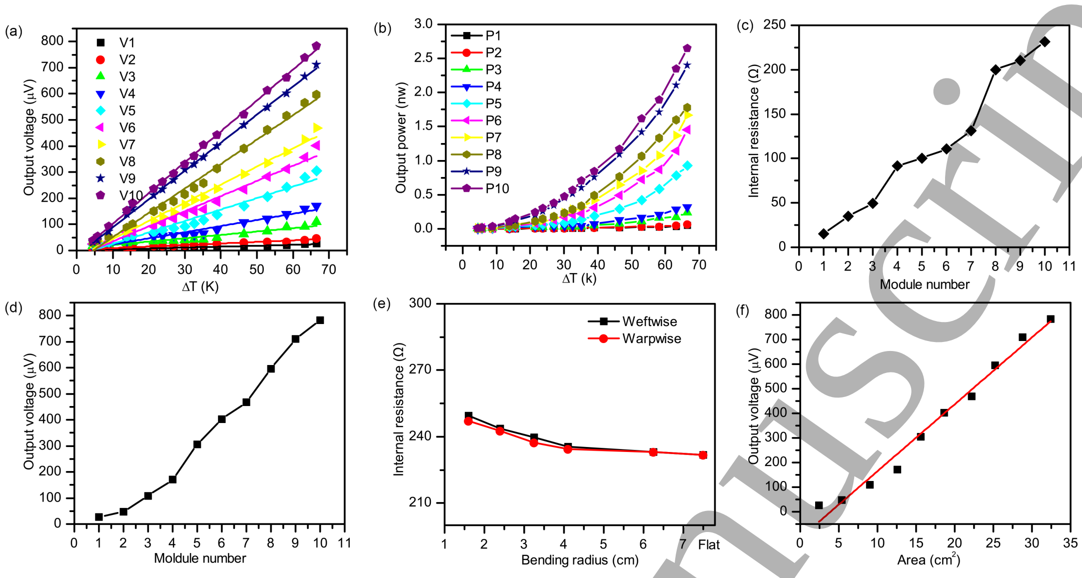

Figure 4. Output thermo-voltage (a) and output power (b) of the fabric TEG with increased temperature difference under different yarn couple modules; Internal resistance (c) and output thermo-voltage (d) with increased yarn couple modules under a fixed temperature difference of 66K; (e) Internal resistance under decreased bending radius at weftwise and warpwise directions; (f) output thermo-voltage per area of the fabric TEG

3.4 Demonstration on body heat conversion

To further demonstrate the possibility of applying the designed fabric TEG for body heat conversion, we adhered the spacer fabric TEG prototype to a human wrist and measured the generated thermal voltage by utilizing the temperature difference between the skin and environment, as shown in Figure 5 (a) and (b). In the initial state, our body was the heat source had a skin temperature of 33.2°C, while the room air had a temperature of 26.8°C. A thermo-voltage of 0.025mV was generated. To enlarge the temperature difference, we cooled the fabric layer exposed to air via iced water. It can be clearly observed that higher thermo-voltage could be generated with the gradually increased temperature gradient. The thermo-voltage reached 0.1242mV under a temperature gradient of 10.7°C. After removed the iced water, the temperature gradient decreased significantly, and the thermo-voltage also decreased correspondingly. This verifies that the designed 3D fabric TEG is feasible in body heat conversion. In addition, an interest phenomenon was found that the fabric TEG appears to generate higher thermo-voltage in temperature difference enlargement procedure than in temperature reduction procedure. The reason for this phenomenon is still not well understood and needs to be studied further. In general, this demonstration verifies that the designed 3D fabric TEG structure is feasible and useful in body heat energy conversion. To further improve the output voltage, more yarn modules should be incorporated into the fabric TEG. It should be noticed that the thermal contact resistance of the fabric TEG to human body and air would cause the real temperature difference along the yarn leg is much smaller than the body to air temperature difference, which will low down the working efficiency of fabric TEG in real wearable situation. Therefore, strategies to reduce the thermal contact resistance are necessary. Since the air between human body and cloth is the major reason for large thermal resistance, one possible solution is to incorporate the fabric TEG with tight-fitting clothing, which could significantly narrow down the air-gap between human body and fabric TEG. In addition, to add a thin layer of thermal grease/tape or to apply a thermal conductive ceramic coating on the surface of fabric TEG would further improve the thermal conduction of fabric TEG to human and air.

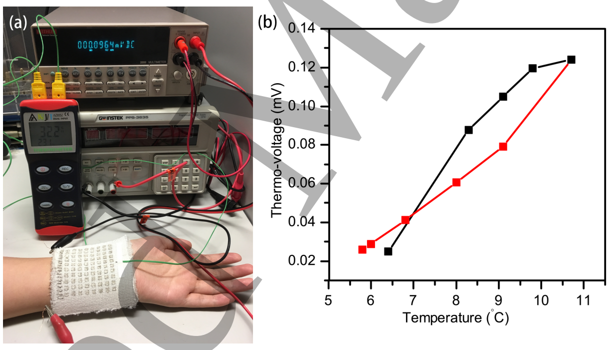

Figure 5. (a) The demonstration of the 3D fabric thermoelectric generator on body heat conversion; (b) Temperature difference-thermovoltage function of the generator prototype, black line and red line indicate the temperature difference enlargement procedure and reduction procedure, respectively. The movie of this demonstration is available at support information S1.

This novel 3D fabric TEG is simple to fabricate without any expensive equipment. It is environmental friendly, cost-effective, controllable, and feasible for large-scale fabrication. Unlike the multi-layered film TEGs reported by other researchers, the 3D fabric TEG is permeable to air and moisture, making the device flexible and comfortable to wear. A temperature difference between the inner and outer surfaces of the clothing can be used for the thermoelectric power generation. The fabric TEG device would have better performance when there is a large temperature difference between the body and environment. It is therefore favored to use in cold weather clothing. Although the efficiency of fabricated prototype is low, this 3D fabric TEG structure design is innovative and provides new thoughts for the study of flexible and wearable TEG. Several strategies will be investigated to increase the power output of the 3D fabric TEG in our future studies, such as using highly efficient inorganic thermoelectric materials, improving the thermal contact between fabric TEG and human skin by incorporating thermal conductive layers, and optimizing the device structures. Besides, although we demonstrate here the concept of using 3D fabric TEG for power generator, the fabric TEG also has potential to be used as temperature sensors.

4. Conclusions

In summary, an innovative wearable 3D fabric TEG structure was designed for the first time and a prototype was fabricated to verify the effectiveness of designed structure. By using a 3D fabric as substrate and thermoelectric yarns as legs, a flexible TEG with sandwich structure can be easily achieved. This fabrication process is simple and easy to operate, which is beneficial to large-scale production. This structure can allow generating temperature difference through the fabric thickness direction as classic inorganic TEGs whilst retaining good flexibility. Hence, it can be applied in wearable situations to convert body heat, especially in some winter wears in which a larger temperature difference between body and air can be achieved. As for now, the spacer fabric TEG prototype only generate a low thermo-voltage of ~800 μV and output power of ~2.6 nW at $\Delta T = 66$ K. To further improve the efficiency of 3D fabric TEG in future, other highly efficient thermoelectric materials such as inorganic thermoelectric materials can be applied, and the conductive connections between yarn legs need to be reinforced. Besides, strategies to improve the thermal contact between TEG and human skin are necessary in real wearable situation, such as using tight cloth and applying ceramic coating. Generally, the 3D fabric TEG structure designed in this study provides a new way for developing flexible and wearable thermoelectric generators.

Acknowledgements

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