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Fabric-based flexible thermoelectric generators: Design methods and prospects

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At the beginning of the 21st century, micro-nano processing technology has developed rapidly, and research on wearable electronic devices has exploded. In particular, flexible thermoelectric materials and thermoelectric generators (TEGs) that have stretchable or foldable properties have been widely studied. Textile-based thermoelectric materials are more suitable for use on the human body than are film or block types of devices, and they are accordingly favoured for wearable smart devices. The design idea of textile thermoelectric materials include the preparation of thermoelectric fibres, textiles, fabric-based films and some novel methods, like using three-dimensional modules to become two-dimensional fabrics by a weaving method, which not to rely something as a base for TE modules. Investigators are thus gaining opportunities to make comfortable and effective fabric TEGs using a wider variety of materials. This article briefly introduces the common methods used to produce textile TEGs. The opportunities and challenges of fabric TEGs are discussed at the end.

KEYWORDS

wearable device, flexible thermoelectric material, thermoelectric generators, fabric, fiber

Introduction

The history of thermoelectric materials spans three centuries (Beretta et al., 2019). Nowadays, with the rapid development of wearable devices, researchers have begun to explore more-flexible thermoelectric generators (TEGs) because rigid TEGs based on inorganic blocks have difficulty in meeting the needs of these applications. Flexible TEGs need to consider wearability, so the design of such a device concerns not only its hotspot performance but also the necessity to make trade-offs between mechanical performance and comfort. Flexible TEG designs include blocks, membranes and textiles. The thermoelectric material used in a block-type TEG is a traditional rigid material that is wrapped in a flexible substrate. Such TEGs produce higher power, which can reach the mW level. Membrane TEGs use thin-film thermoelectric materials attached to a flexible base plate. The advantage of a membrane TEG is that its structure has design flexibility.

Both *n*-type and *p*-type thermoelectric films can be used in a variety of design methods to accommodate bending, folding, cross-tiling and so on. These methods are all attempts to make the hotspot module as three-dimensional as possible for the environment in which it is to be used, in order to obtain higher energy efficiency than is possible with a two-dimensional surface (Yamamoto and Takai, 2002; Snyder et al., 2003; Lee et al., 2016; Ito et al., 2017; Mather and Wilson, 2017; Wu and Hu, 2017; Nan et al., 2018; Jia et al., 2019). Textile TEGs use fabric as a substrate on which to integrate thermoelectric fabrics or thermoelectric films. The common methods for producing textile TEGs include wet textiles, drip or dip coating, screen printing and magnetron sputtering. In addition, a three-dimensional weaving method has recently been introduced in which a single thermoelectric module is made into a bundle-like collection and then wrapped. The bundle of modules is then woven into a fabric, and the core of the fibre is left exposed at the junction used for connecting the thermoelectric module. This three-dimensional weaving method does not need to rely on a substrate to produce a fabric-type TEG.

Development of thermoelectric materials

In the first half of the 19th century, Ørsted coined the term 'thermoelectric', and he created the first thermoelectric pile, made of bismuth and antimony (Roget, 1832), with the help of the mathematician and physicist Jean Baptiste Joseph Fourier. In the middle and second half of the 19th century, the three basic thermoelectric effects were well explained (Thomson, 2017), and heat-source power-generation devices using metallic conductors were developed (Kennedy, 1903); they were the first TEGs. At that time, thermoelectric materials were not very efficient, and people have been struggling ever since to find classes of materials with good thermoelectric properties. In the middle of the 20th century, Ioffe proposed a theory of the thermoelectric effect in semiconductors, which greatly increased the range of potential thermoelectric materials (Beretta et al., 2019). He also introduced the dimensionless thermocouple figure of merit, ZT (Ioffe, 1957). After semiconductor materials began to be used to make TEGs, the ZT value increased from the 19th-century value ZT = 0.1 to ZT = 1.0. At that time, people began to recognise the potential benefits of thermoelectric materials and to pay attention to them. In the 1980s, researchers prepared a variety of wearable devices (Mann, 1997), including computers, cameras and sensors. Without exception, none of them looked aesthetically pleasing and comfortable, and some even looked bulky because they had to be plugged into huge power supplies. Today, however, the development of micro-nano processing technology has led to their viability in many fields, materials, electronics and biology-and microelectronics and micro-power sources have also been created. The ability to modify thermoelectric materials has also long been upgraded to the microscopic level (Chen et al., 2019; Chen et al., 2020; Chen et al., 2021). The exploration of wearable TEGs has never stopped, and people can see the prospects and hopes for future wearable smart devices in products such as Google Project Glass, Ralph Lauren's smart jersey and OM Signal.

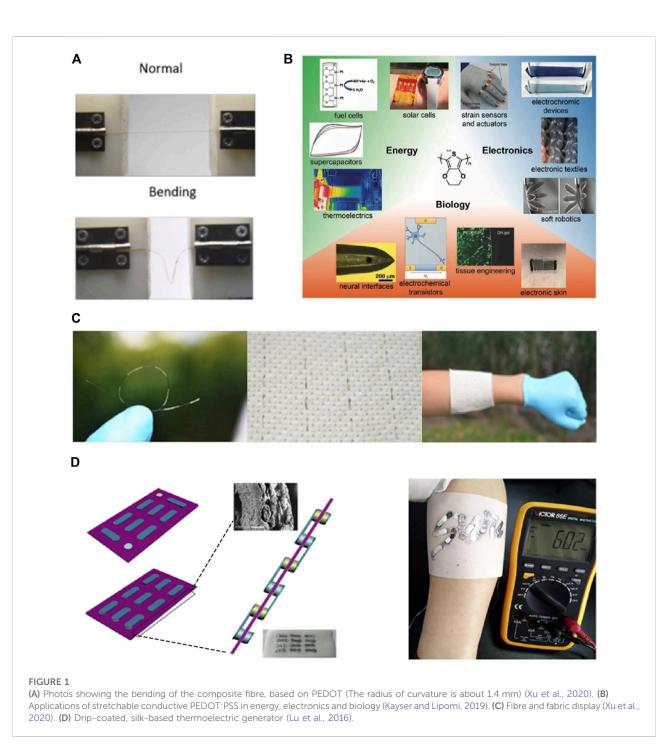
Fabric thermoelectric generators

As noted above, the common methods for producing fabricbased thermoelectric material technologies include wet textiles, drip or dip coatings, screen printing and magnetron sputtering. 'Wet textiles' refer to the original step of spinning a stock solution doped with materials that have thermoelectric properties, such as carbon poly nanotubes (CNTs) and (3, 4ethylenedioxythiophene) (PEDOT) complex with poly (styrene sulfonate) (PEDOT:PSS). So, far as possible, they are dispersed evenly in a solution, and the mixture is sprayed from a hole into a pool for solidification, after which it can be washed and dried to produce *n*-type or *p*-type spinnable thermoelectric fibres. These fibres can be modified by using different doping and different stock solutions. However, the thermoelectric properties of the resulting fabrics are generally not as high as those of other types of thermoelectric materials. Because PEDOT:PSS is a polymer with a high electrical conductivity σ , which also has good tensile strength and flexibility (Xu et al., 2020) (Figures 1A,C), it is a very good sheet-conductive material, which is often used in flexible and stretchable devices (Kayser and Lipomi, 2019) (Figure 1B).

At present, PEDOT:PSS can be made into fibres by wet weaving, which can achieve a conductivity $\sigma = 4,039 \pm 215 \text{ S cm}^{-1}$, a Young's modulus of 22 GPa, and a fracture stress of 550 MPa, which is also the highest value reported for PEDOT to date (Sarabia-Riquelme et al., 2020).

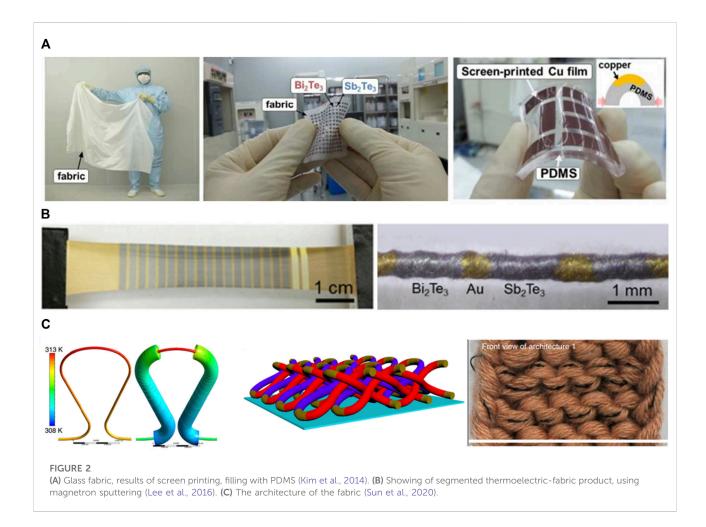
(Xu et al., 2020) have used PEDOT:PSS/Te to prepare TE fibres, which they have then woven into TE fabrics. They added Te nanowires, Polyethylene glycol (PEG Molecular Weight ~400), PEDOT:PSS pellets and other substances to make the original solution, which they then dispersed uniformly through a syringe into the isopropyl alcohol coagulation bath. After spinning, the resulting fibres were dried and repeatedly coated with silver paste to obtain the required thermoelectric composite fibres, as shown in Figures 1A,C.

Drip or dip coating is a method for depositing a thermoelectric film on a fabric base, and it has good applicability. First, a solution containing a thermoelectric material is prepared, and it is then dripped onto a substrate, infiltrates the fabric substrate and is finally dried. This deposition process can be performed repeatedly. This preparation method is simple, but the binding strength between the film and substrate is not high, and the film thickness is difficult to control (Lu et al., 2016) produced a silk fabric TEG from a paste of the raw materials Te, Bi and CNTs, which they repeatedly deposited on a substrate of *Bombyx mori* cocoons to make a textile-based TEG (Figure 1D).



Screen printing is also a common method for preparing thermoelectric fabrics. Kim et al. (2014) synthesised *n*-type Bi_2Te_3 and *p*-type Sb_2Te_3 and printed them onto a glass fabric using a screen-printing technique. They placed the resulting Bi_2Te_3 and Sb_2Te_3 films in ambient N₂ at 530°C for 10 min and at 500°C for 30 min, respectively. Under mechanical pressure, the films crystallised and densified. Finally, the investigators embedded copper foil into polydimethylsiloxane (PDMS) to form the top and bottom plates, stacked them together and immersed them in the PDMS to fill the voids (Figure 2A).

Magnetron sputtering is another common film-preparation method, the coating repeatability is high, the substrate requirements are low and the resulting film is compact and highly binding (Lee et al., 2016) devised a method using magnetron-sputtering technology with high-purity Bi_2Te_3 and



Sb₂Te₃ targets for producing thermoelectric fabrics. After spinning the substrate and depositing TE membranes on polyacrylonitrile (PAN) nanofiber plates with a density of about 0.15 mg cm⁻². They deposited Bi₂Te₃ and Sb₂Te₃ to prepare separate *n*-type and *p*-type yarns, which they twisted and wound to make segmented thermoelectric fabrics (Figure 2B).

Each of the above four methods has its limitations. The thermoelectric properties of the fabrics from 'wet textiles' method are generally not as high as those of other types of thermoelectric materials. For drip or dip coating method, the binding strength between the film and substrate is not high, and the film thickness is difficult to control. Screen printing requires high temperature and mechanical pressure to make the films crystallised and densified. Magnetron sputtering have to synthesize n-type and p-type yarns separately then make segmented thermoelectric fabrics.

Recently, (Sun et al., 2020) introduced the novel method of weaving a thermoelectric module into a fabric. They designed a threedimensional thermoelectric module using the interlock-weaving method to form a two-dimensional structure. They obtained an output voltage from this structure that is 24 times greater than that obtainable from a two-dimensional architecture. Based on basic thermal theories (Priya and Inman, 2009; Nan et al., 2018) (Figure 2C), the collection of human heat and the dissipation of cold-end heat are essential for a high-efficiency TEG. This Sun et al. device not only can make good use of the temperature gradient—like a three-dimensional thermoelectric module—but also fits the human body like a fabric. Therefore, unlike a TEG, which requires a substrate to support its thermoelectric module, the mechanical properties, stability and power from this device are not affected by the substrate. It has a high output power—a power density of 70 mW m⁻² at 44 K—and has excellent stretchability. It has a biaxial stretchable property rather than being stretchable only uniaxially. The output voltage fluctuates by only 4% during elbow movement, so it is almost unaffected by the movement and has fairly good practicality.

Challenges and opportunities

Fabric-based TEGs are produced using a variety of methods, all of which are designed to solve the problem of flexibility and refinement of the thermoelectric materials. For fabric-based thermoelectric materials and TEGs, oxidation, mechanical life and losses are always issues to be overcome. Discovering new structural and functional materials and proposing novel and more efficient structural designs are clearly useful directions to explore in order to improve the practicality of textile TEGs. In addition, the successful development of flexible inorganic thermoelectric materials in 2019 has provided a new direction for the development of flexible thermoelectric materials (Liang et al., 2019). More preparation processes and design methods for TEGs thus remain to be explored.

Author contributions

All authors listed have made a substantial, direct, and intellectual contribution to the work, and approved it for publication.

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Conflict of interest

The reviewer CH is currently organizing a Research Topic with the author XC.

The remaining authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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