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1 A comprehensive review on the output voltage/power of wearable
2 thermoelectric generators concerning their geometry and thermoelectric
3 materials
4

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8

9 **Abstract:** Wearable thermoelectric generators (TEGs) are considered as a promising power supply for
10 low power wearable electronics. To obtain high thermoelectric (TE) generation, the focus should be
11 on two main factors, including TE materials and the configurations of TE legs. Concerning these two
12 factors, this paper provides a comprehensive review of recent studies on wearable TEGs. In general,
13 TE materials can be classified into three categories, including inorganic, organic, and hybrid
14 (inorganic-organic). In addition, the TE legs can be prepared in three different configurations,
15 including ingot-shaped, film-shaped, and yarn-shaped. Based on the reviewed literatures, the superior
16 output powers of all the three configurations were achieved by the inorganic, hybrid, and organic TE
17 materials, respectively. It should be noted that the ingot- and the yarn- shaped legs were mostly
18 composed of the inorganic and the organic TE materials, respectively. Whereas, all the three types of
19 TE materials were almost equally used to prepare the film-shaped legs. Regarding power density, the
20 ingot-shaped legs stood first followed by the film- and the yarn- shaped legs, respectively. Precisely,
21 the output powers of the ingot- and the film- shaped legs were at $\mu\text{W}/\text{cm}^2$ level, dropping to nW/cm^2
22 for the yarn-shaped legs.

23
24 **Keywords:** Energy harvesting, wearable thermoelectric generators, power density, configuration of
25 thermoelectric legs, thermoelectric materials.
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Nomenclature

Symbols

α	Seebeck coefficient ($\mu\text{V/K}$)
σ	electrical conductivity (S/cm)
λ	thermal conductivity (W/mK)
$Z\bar{T}$	figure of merit
T_H	temperature of hot reservoirs
T_C	temperature of cold reservoirs
A	cross-sectional area (cm^2)

Acronyms

CNTs	carbon nanotubes
CNF	Cellulose nanofiber
CNTFs	carbon nanotube fibres
FPCB	flexible printed circuit board
MWCNT	multi-walled carbon nanotubes
PET	polyethylene terephthalate
PEI	polyethylenimine
PEDOT: PSS	poly(3,4-ethylenedioxythiophene)
	polystyrene sulfonate
PDMS	polydimethylsiloxane
PCM	phase change material
SWCNT	single-walled carbon nanotubes
TEG	thermoelectric generator
TE	thermoelectric module

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52 1. Introduction

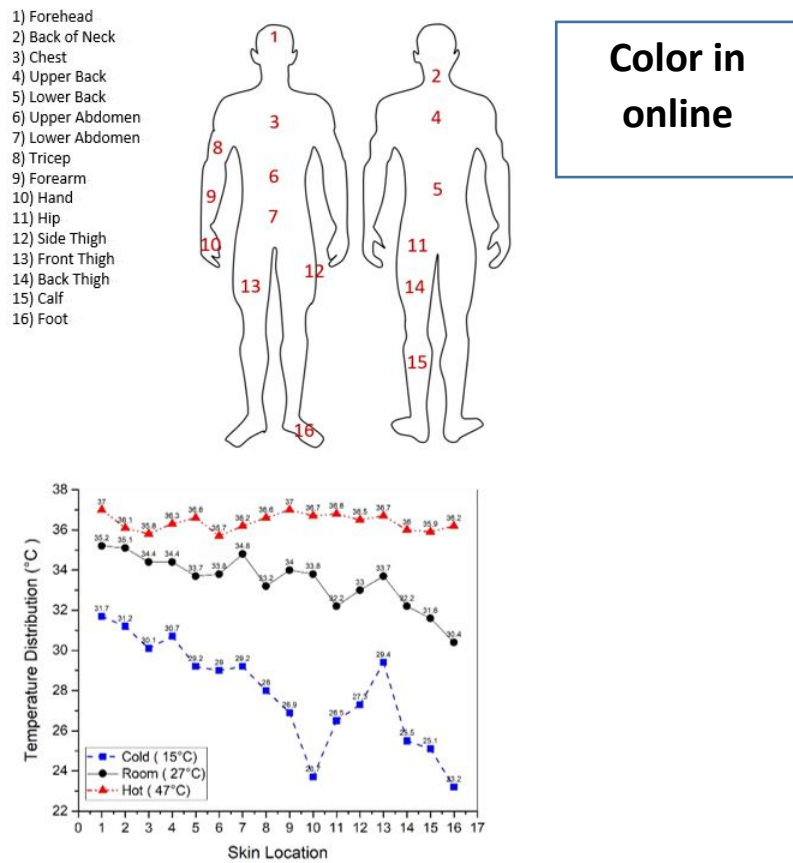
53
 54 Progress in wearable technologies such as activity trackers and portable health monitors has emerged
 55 considerable interests in lightweight, durable, and on-board power supplies [1]. Thus, harvesting the
 56 available energies in the daily environments has become a promising candidate. In general, there are
 57 two approaches to scavenging energies for wearable electronics, including active and passive [2]. The
 58 active approach is based on wireless power transmittance, sending a wireless power from one or more
 59 external power sources to wearable devices [3]. Thus, this approach can maintain normal device
 60 operation in long-term experiments [4]. However, its main drawback is limiting the wearers' movement
 61 to a specific distance from the external power source [5]. Regarding the passive approach, human body
 62 is used as the energy supplier. In general, the energy sources of human body include kinetic energy [6-
 63 8], thermal energy [9-10], chemical energy [6] [11], and respiration [12-13]. As Table 1 shows, there
 64 are specific energy harvesters to convert these energies into electrical energy.

65 **Table 1.** An overview of potential energy sources of the human body and their associated energy harvesters.
 66

Energy Type	Energy Harvester Subcategories	Ref	
Biochemical Energy	Biofuel cells	Microbial Fuel Cells [14], [15], [16]	
		Enzymatic Fuel Cells [17], [18], [19]	
Thermal Energy	Thermoelectric	[20], [21]	
	Pyroelectric	[22], [23]	
Kinetic Energy	Piezoelectric	[24], [25], [26], [27]	
	Triboelectric	[24], [25], [28], [29]	
	Electromagnetic	Inertial induction	[25], [30]
		Gear-and-generator	[31], [32]
Exhaled Air Energy	Triboelectric	[33], [34]	
	Piezoelectric	[35]	
	Pyroelectric	[36]	
	Thermoelectric	[37]	

67
 68 As a warm blooded species, human body continuously generates thermal energy in a range between
 69 100W and 525W [38]. For example, as Fig. 1 shows, body temperature varies in a range of between
 70 23°C and 37°C when room temperature fluctuates between 15°C and 47°C [39]. Capturing the
 71 dissipated heat from human body with TEGs (with ~1% conversion efficiency) generates ~1W to
 72 5.25W electrical power. Although this amount of energy is sufficient to power a vast range of wearable
 73 sensors, it requires covering the whole body with TEGs, which is impractical [7]. In addition, achieving

74 a wearable TEG with high conversion efficiency (~1%) depends on several critical factors, including
 75 the thermoelectric (TE) materials [40], the geometry of the TE legs [39, 41-42], the structure of
 76 heatsinks [43-45], the metabolic rate of wearers (e.g. resting, walking or running state) [46], the
 77 mounting position of the TEGs (e.g. wrist, chest, arm) [46-47], and ambient thermal conditions (e.g.
 78 dry bulb temperature, relative humidity, and air velocity) [48]. Therefore, precise considerations are
 79 required at the design stage of wearable TEGs, because any deficiency at this stage impacts the output
 80 power of the TEGs.



81
 82 **Fig. 1.** Variations in the temperature of body parts at different ambient temperatures, adapted from [39].

83
 84 Hence, this study aims to thoroughly review the recent studies on wearable TEGs concerning the
 85 configuration and materials of their TE legs. Precisely, herein, wearable TEGs are categorized based
 86 on the configuration and materials of their TE legs to provide a reliable comparison between their
 87 output voltages/powers. This is significantly important because, several excellent reviews have been
 88 conducted recently on wearable TEGs with a specific focus on either the TE materials [49-54] or a
 89 specific configuration of TE legs [9, 38, 55-57]. Nevertheless, to the best of our knowledge, so far no

90 study has considered the impacts of both factors (i.e. the configurations and materials of the TE legs)
91 on the output voltage/power of wearable TEGs at the same time. However, since both factors have
92 direct impacts on the output voltage/ power of wearable TEGs, it is not reliable to compare the
93 performances of the TEGs with each other concerning only one factor.

94 Accordingly, the paper is organized as follows. First, the theoretical basis of TEGs is summarized in
95 Section 2, including different configurations and materials of wearable TEGs. Section 3 reviews
96 wearable TEGs comprising of ingot-shaped legs with regard to their TE materials. Likewise, Sections
97 4 and 5 respectively review the recent researches on film-shaped and yarn-shaped legs regarding their
98 TE materials. Section 6 explains some challenges and future trends. Section 7 summarises the main
99 outcomes of this review.

100

101 2. Theoretical principle of TEGs

102

103 Thermoelectric generators (TEGs) are solid-state semiconductor devices that convert waste heat
104 directly into electrical energy. The TE theories are based on the Seebeck effect, which was discovered
105 in 1821 by a German physicist called Thomas Johann Seebeck. Based on the Seebeck effect, an
106 electrical power is generated when a temperature gradient exists between the two ends of a
107 thermocouple. To illustrate, a thermocouple consists of two electrically different conductors or
108 semiconductors that are directly in contact or electrically connected together, allowing the charge
109 carriers flow between the materials. Thus, when a temperature difference occurs between the two ends
110 of a thermocouple, the charge carriers diffuse away from the warmer side of the thermocouple and
111 build up at the cooler end. This movement results in more charge carriers at the colder part than the
112 warmer part, leading to the generation of an open circuit voltage called the Seebeck voltage (V).
113 Following Equation 1, the Seebeck voltage is directly related to the temperature difference (ΔT) along
114 the thermocouple by a proportional factor called the Seebeck coefficient (α).

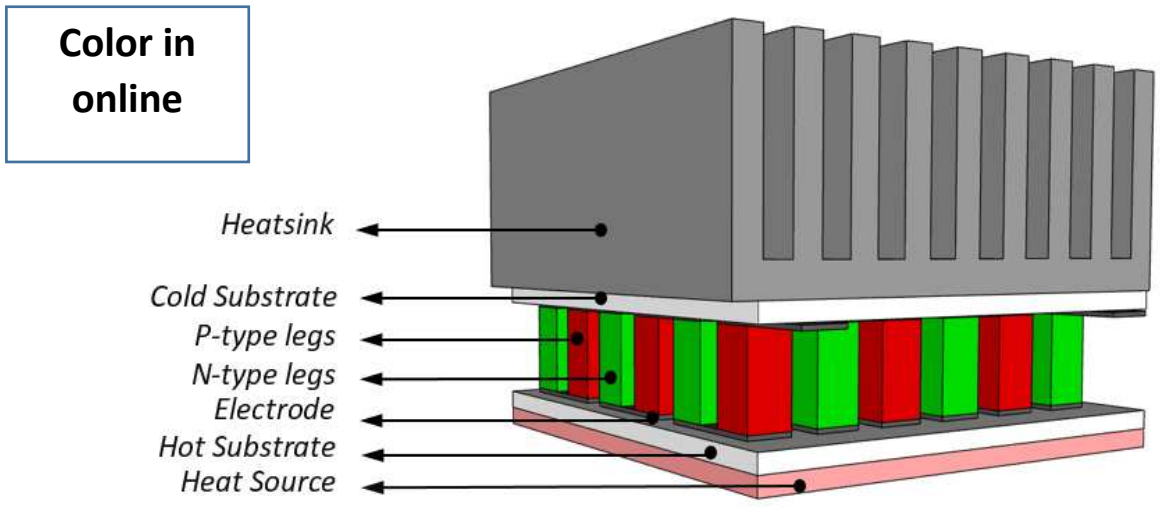
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$$116 \quad V = \alpha \Delta T \quad (1)$$

117

118 Accordingly, a wearable TEG consists of an array of p - and n -type semiconducting legs comprising
119 surplus holes and electrons, respectively. Using metal electrodes/interconnects, the TE legs (i.e. the

120 semiconductors) are connected electrically in series and thermally in parallel to form thermocouples.
 121 A conventional TEG is composed of a large number of thermocouples sandwiched between two
 122 electrically insulating and thermally conductive substrates. A temperature difference between the
 123 substrates leads to a temperature gradient along the legs. Thus, the free charges (i.e. electrons and
 124 holes) in the legs start moving from the hot side to the cold side, converting the thermal energy into
 125 the electrical energy. In general, higher output voltage/power can be achieved by increasing the
 126 temperature gradient in the thickness direction of the legs. Maintaining an adequate temperature
 127 difference along the legs, a heatsink is usually attached to the cold side of the TEGs to accelerate the
 128 heat dissipation [58]. Fig. 2 depicts the standard configuration of a conventional TEG.



129
 130 **Fig. 2.** A schematic diagram of a typical wearable TEG and its components.

131
 132 In general, the energy conversion efficiency of a TEG depends on the TE properties of its constituent
 133 legs' materials and the temperature difference along the legs [51]. Accordingly, the maximum
 134 theoretical conversion efficiency (η_{\max}) of a TEG can be expressed by [57]:

135
$$\eta_{\max} = \left(\frac{T_H - T_C}{T_H}\right) \cdot \left(\frac{\sqrt{1 + Z\bar{T}} - 1}{\sqrt{1 + Z\bar{T}} + \frac{T_C}{T_H}}\right) \quad (2)$$

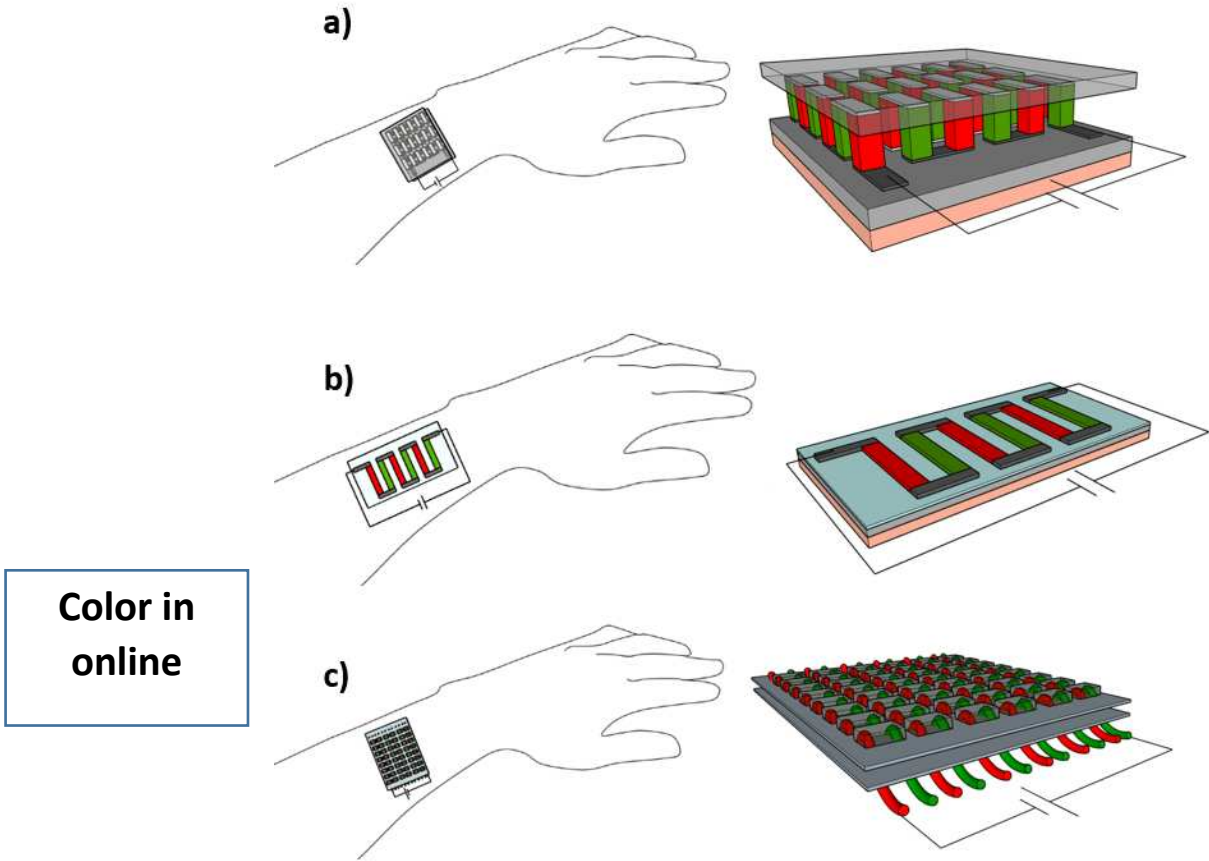
136 where T_C and T_H are the cold- and hot-side temperatures, respectively, and $Z\bar{T}$ is the figure of merit of
 137 the TE materials. The term in the left parenthesis is called Carnot efficiency (η_C), which is an upper
 138 limit on using the waste heat for TE power generation [51]. According to Carnot efficiency (η_C), the
 139 conversion efficiency (η_{\max}) of TEGs is directly related to the temperature difference between the hot

140 and cold junctions of the TE legs. To specify, increasing the temperature difference along the legs rises
141 the conversion efficiency of the TEGs. To tune the Carnot efficiency (η_C), one of the most promising
142 approach is manipulating the structural design of the TE legs respective to the optimal ratio of
143 the cross-sectional area of the p -type (A_p) and n -type (A_n) legs. The optimal ratio could be measured
144 by:

$$145 \quad \left[\frac{A_n}{A_p} \right] = \sqrt{\frac{\sigma_p \lambda_p}{\sigma_n \lambda_n}} \quad (3)$$

146
147 where σ_p and σ_n are the electrical conductivity (S/cm) and λ_p and λ_n are the thermal conductivity
148 (W/m K) of the p - and n -type materials, respectively. Regarding the cross-sectional area of the legs,
149 their configurations can be classified into two main categories: transversal (cross-plane) and lateral
150 (in-plane). This classification is related to the manner of arranging the legs on the substrates and
151 subsequently to the direction of the temperature gradient within them [59]. Regarding the cross-plane
152 design, the TE legs are perpendicular to the hot (i.e. the skin) and cold (i.e. the environment) sources,
153 as Fig. 3a shows. Notably, the temperature difference between the human body and the environment
154 results in a perpendicular heat flux direction to the skin. Therefore, the temperature difference in the
155 direction of the legs' thickness encourages the free holes and electrons in the legs to move vertically
156 and generate electricity. However, it is desirable for a wearable TEG to be lightweight and flexible to
157 take on the arbitrary shape of the body's surfaces (e.g. wrist) [60]. Conversely, the cross-plane design
158 suffers from rigidity and bulkiness due to the high thickness of the legs, causing cumbersome for the
159 wearers. Thus, the most promising alternative to solve this issue is the in-plane design, consisting of
160 two dimensional (2D) film-shaped thermocouples fabricated onto a supporting substrate and in parallel
161 with the skin, see Fig. 3b [61]. Thus, splitting the substrate in half either from the length or width, the
162 opposite halves serve as the hot and cold substrates. As a result, the film-shaped legs merely harvest
163 the temperature gradient in the length/width direction and rarely capture the desirable temperature
164 difference in the thickness direction. Accordingly, the in-plane design usually suffers from lower
165 conversion efficiency than the cross-plane one [62-63]. To solve the rigidity and efficiency issues at
166 the same time, researchers have proposed to combine these two designs together, resulting in the yarn-
167 shaped thermocouples. As Fig. 3c depicts, 2D TE yarns are knitted together or sewn onto a three

168 dimensional (3D) or 2D flexible substrate, allowing the flexible yarns to utilize the temperature
 169 difference in the thickness direction [64].



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 171 **Fig. 3.** Schematics of different configurations of thermocouples in wearable TEGs: a) ingot-shaped; b) film-shaped; and
 172 c) yarn-shaped.

173
 174 Apart from the configuration of the TE legs, it is also of critical importance to generate them with high
 175 $Z\bar{T}$ value materials. To specify, $Z\bar{T}$ value expresses the heat-to-power conversion efficiency of materials
 176 and is measured by:

177
$$Z\bar{T} = \frac{(\alpha^2 \sigma T)}{\lambda} \tag{4}$$

178 where α is the Seebeck coefficient ($\mu\text{V/K}$), σ is the electrical conductivity (S/cm), T is the absolute
 179 temperature (K), and λ is the thermal conductivity (W/m K) of the materials. Notably, the numerator,
 180 $\alpha^2 \sigma$, is called the power factor (PF). To obtain a high $Z\bar{T}$ value material, it is required to increase the
 181 Seebeck coefficient and the electrical conductivity but reduce the thermal conductivity. Designing such

182 a material is challenging because these three TE properties are interdependent, thus it is impractical to
183 individually tune one property without affecting the others [51]. To clarify, although increasing the
184 carrier concentration improves the electrical conductivity of TE materials, it also undesirably decreases
185 and increases their Seebeck coefficient and thermal conductivity, respectively [52]. In fact, the
186 constituent raw materials of the TE materials have direct impact on their TE properties (i.e. Seebeck
187 coefficient, electrical conductivity, and thermal conductivity). To illustrate, TE materials can be
188 classified into three categories, including inorganic, organic, and hybrid (inorganic-organic). The
189 inorganic type consists inorganic elements, such as bismuth telluride (Bi_2Te_3) and its alloys, Lead
190 telluride (PbTe) and its alloys, silicon-germanium (SiGe) alloys, antimony telluride (Sb_2Te_3), and tin
191 selenide (SnSe). This type benefits from high electrical conductivity but suffers from rigidity, scarcity,
192 and toxicity [65]. In addition, based on the life cycle impact assessment of Soleimani et al. [66], it is
193 proved that the inorganic type causes significantly greater environmental impacts than the other two
194 types due to its extremely energy-intensive manufacturing processes. Accordingly, there is a growing
195 interest to switch from the inorganic TE materials to the organic ones, such as carbon nanotubes,
196 graphene, and conductive/non-conductive polymers. Although the organic type privileges low thermal
197 conductivity, its electrical conductivity and Seebeck coefficient are by far lower than those of the
198 inorganic one [52]. Accordingly, the inorganic and organic types are combined together to benefit from
199 the high power factor of the former and the low thermal conductivity of the latter at the same time. The
200 result is called the hybrid type, and it mostly consists of an inorganic material (e.g. metal-based
201 chemicals) as an additive doped into an organic material as the matrix. For example, Lei et al. [67]
202 developed a hybrid TE material by coating $\text{MgAg}_{0.97}\text{Sb}_{0.99}$ composites with multi-walled carbon
203 nanotubes (CNTs) as the inorganic and organic parts, respectively. However, still the hybrid type
204 suffers from two deficiencies, such as consisting toxic and rare inorganic elements in addition to lower
205 conversion efficiency than the inorganic type [52].

206 Therefore, in accordance with Equation 2, the configuration of the TE legs and the $Z\bar{T}$ value of their
207 constituent material directly impact on the conversion efficiency of the TEGs [68]. Accordingly, this
208 study aims to comprehensively review the recent studies on wearable TEGs concerning the
209 configuration and materials of their thermocouples. Ultimately, the output powers/voltages of the
210 reviewed studies were compared with each other to find out the contribution of these two factors to the
211 TE performance of the TEGs.

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3. Ingot-shaped thermocouples

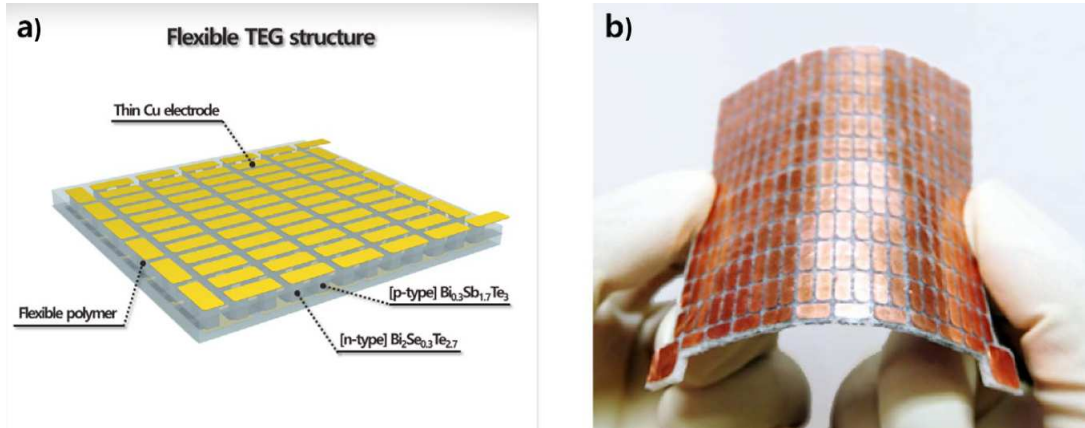
3.1. Inorganic TE materials

Using ingot-shaped thermocouples can be traced back to the generation of very conventional TEGs, when TE mixtures were synthesised via ball milling or melting/heating the TE powders [69]. To obtain the ingot-shaped legs, the as-prepared TE mixtures were consolidated into ingots via cold or hot pressing [70]. Next, the generated TE ingots were bonded together with electrical shunts, resulting in the ingot-shaped thermocouples. Regarding wearable TEGs, their ingot-shaped legs are mostly consisting of Bi₂Te₃-based alloys. In particular, their *p*-type legs mostly comprised of either Bi₂Te₃ or Bi_{0.3}Sb_{1.7}Te₃ composites, while either Bi₂Te₃ or Bi₂Te_{2.7}Se_{0.3} composites are used in the generation of the *n*-type legs. The superiority of these inorganic TE materials is attributed to their dominant energy conversion efficiencies [52]. It should be noted that the ingot-shaped thermocouples could be integrated together using either a flexible or a rigid substrate. However, following recent progresses in wearable TEGs, using flexible substrates such as polymers, printed circuit board (FPCB), holders, and fabrics have drawn growing attentions compared with the conventional rigid (Aluminum Oxide-based) ones. For example, Kim et al. [71] generated a flexible wearable TEG via encapsulating 72 couples of TE legs in a flexible polymer, as Fig. 4 shows. To specify, the *p*-type (Bi_{0.3}Sb_{1.7}Te₃) and *n*-type (Bi₂Sb_{0.3}Te_{2.7}) ingot-shaped legs were connected together with copper strips as electrodes. To hold the legs in place, the gaps between them were filled with a very low thermal conductivity (0.03 W m⁻¹K⁻²) polymer. The authors specified different variables for tuning the output power of the fabricated TEG, such as varying the legs' height (i.e. 0.8 mm and 2.5 mm), the fill factor (i.e. 15.1%, 19.8%, and 27.2%), and the air velocity (i.e. 0 to 3m/s with 1m/s interval). The results showed that increasing the legs' height from 0.8 mm to 2.5 mm raised the power density by four times. Furthermore, the authors demonstrated that a higher fill factor (the ratio of the area covered by the TE legs to the surface area of the TE module) is more efficient at higher air velocities. The authors demonstrated that at 8.9°C temperature difference and zero air velocity, the maximum obtained output power was 2.5 μW/cm² that was achieved by 15.1% fill factor.

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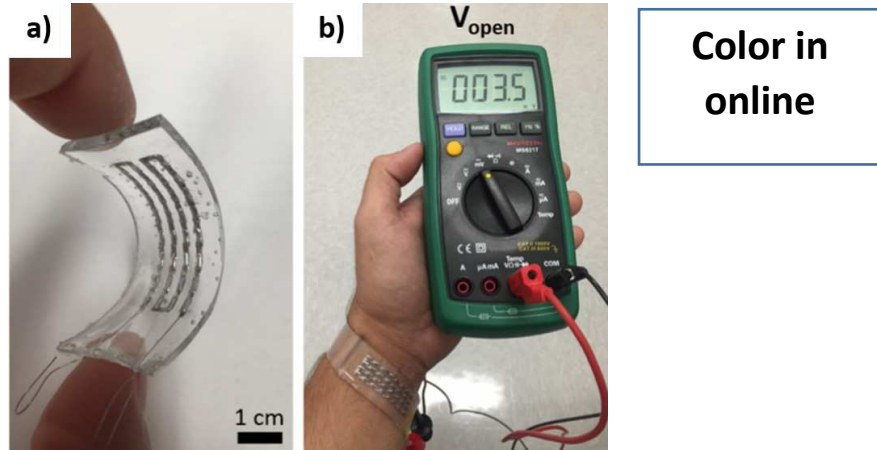
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Fig. 4. a) Schematic of the flexible TEG; b) Photo of the actual fabricated TEG [71].

245 Likewise, Suarez et al. [72] used one of the most common flexible polymer called
246 polydimethylsiloxane (PDMS) to serve as the flexible substrate, see Fig. 5. In particular, the
247 thermocouples consisted of *p*-type (Bi_{0.5}Sb_{1.5}Te₃) and *n*-type (Bi₂Se_{0.3}Te_{2.7}) ingot-shaped legs were
248 electrically connected together using EGaIn. When the TEG comprised of 32 pairs of legs was worn
249 on the wrist (~37°C) at 24°C ambient temperature, the results showed that the maximum recorded
250 output power was 1 μW/cm². However, the authors demonstrated that using a porous polymer-based
251 substrate was superior to the solid one. To illustrate, the performance of the developed TEG was
252 compared with that of a conventional rigid TEG surrounded with air. The results proved that fully
253 coating the legs with the PDMS layer led to a lower output voltage and power than a conventional rigid
254 TEG. This was mainly because the PDMS layer with 0.15W/mK thermal conductivity served as a
255 thermal pass compared with the surrounding air layer (0.025 W/m.K) in the rigid TEGs. Therefore, the
256 authors proposed using a porous PDMS substrate to decrease its thermal conductivity down to 0.08
257 W/mK, which resulted in 86% greater output voltage.

258

259



260
 261 **Fig. 5.** a) Photo of the fabricated TEG; b) The open circuit voltage of the fabricated TEG at room temperature [72].

262 Regarding the negative effect of the polymer substrate on the output power of the TEGs, Jung et al.
 263 [73] attempted to reduce the thermal conductivity of the PDMS substrate by producing it spongy. To
 264 clarify, the authors encapsulated the legs in a PDMS sponge (0.08 W/mK) possessing a lower thermal
 265 conductivity than a plain PDMS substrate. To prepare the sponge, the authors developed sugar template
 266 by pressing granule-sized sugar (~300 μm) in a stainless-steel mold for 60 seconds. Then the prepared
 267 sugar template was dipped into the plain PDMS mixture. After degassing the mixture, the PDMS-
 268 coated template was put in deionized water and stirred thoroughly at 100°C for 5hrs, leading to the
 269 generation of the PDMS sponge. Next, the as-prepared PDMS sponge was inserted into 47 Bi₂Te₃–
 270 based thermocouples each with the dimensions of 1.2mm × 1.2mm × 2.6mm, as Fig. 6 illustrates.
 271 Finally, the thermocouples were connected together using copper-foils as electrodes. The results
 272 demonstrated that the fabricated TEG generated nearly 130 μW/cm² at 8°C temperature difference.

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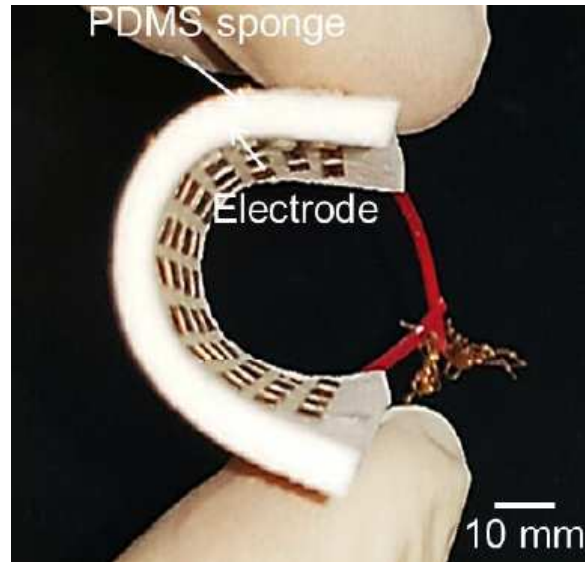


Fig. 6. Photo of the actual fabricated TEG [73].

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275
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277 To further diminish the negative effect of coating the legs with a polymer-based substrate, Hong et al.
278 [74] coated only a small part of the legs with a commercial flexible polymer called Ecoflex. To specify,
279 24 thermocouples were prepared using $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ (*p*-type) and $\text{Bi}_2\text{Te}_{2.7}\text{Se}_{0.3}$ (*n*-type) composites,
280 while they were electrically connected together by copper strips as electrodes. Then, Ecoflex was
281 poured at the two ends of the thermocouples to form two separate 1mm thick substrates. It should be
282 noted that only 0.5mm of each end was coated with the flexible substrate, leaving the rest height (i.e.
283 4mm) of the legs exposed to the ambient air, see Fig. 7. In addition, the authors attempted to improve
284 the lateral thermal conductivity of the Ecoflex layer by adding aluminium nitride micro particles to it.
285 The results revealed that at ΔT of 10°C , the fabricated TEG achieved the highest output powers of 4.5
286 $\mu\text{W}/\text{cm}^2$ and $10.7 \mu\text{W}/\text{cm}^2$ under the sitting and walking conditions, respectively.

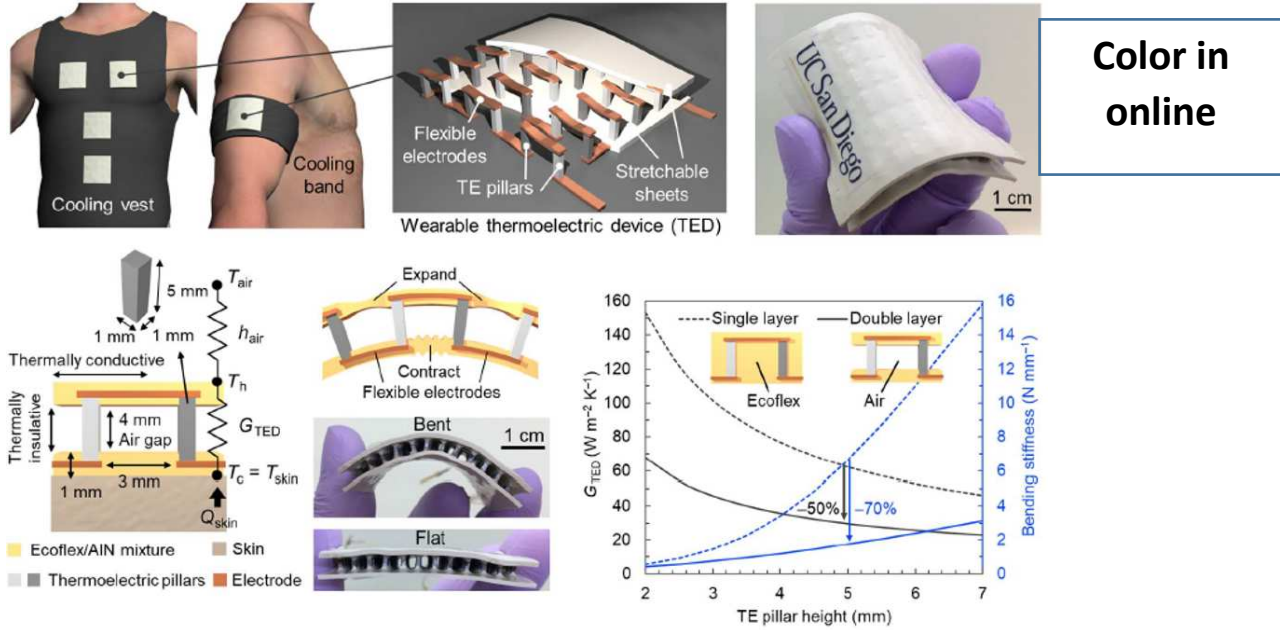
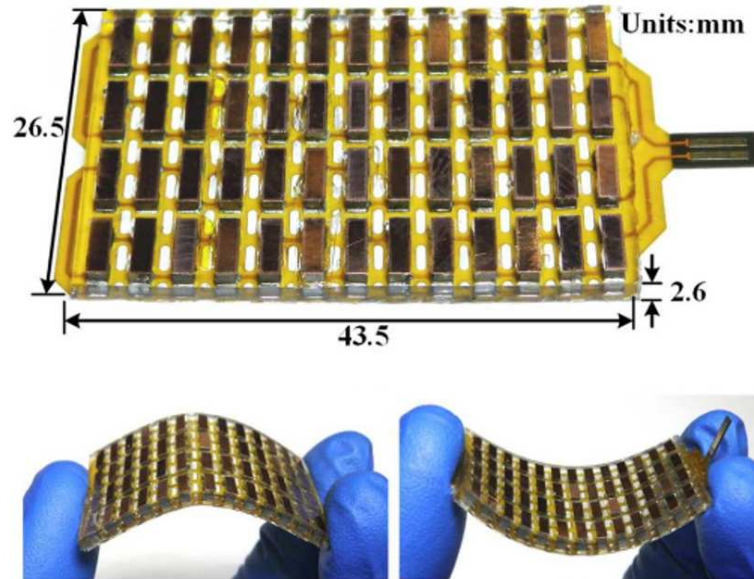


Fig. 7. Internal and external configurations of the developed TEG [74].

Apart from applying a flexible substrate, Wang et al. [75] focused on improving the flexibility of the TEG via replacing the rigid electrodes with the flexible ones. Accordingly, the authors replaced the bottom copper strips with a flexible printed circuit board (FPCB). To specify, 52 pairs of p -($\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$) and n -($\text{Bi}_2\text{Se}_{0.5}\text{Te}_{2.5}$) type legs were soldered onto a FPCB from the bottom, as Fig. 8 shows. Notably, several holes were cut in the FPCB to further increase its bendability. Then, the legs were soldered from the top (i.e. the cold junctions) onto copper strips as the top electrodes. Holding the legs in place, the fabricated TEG was immersed in a PDMS mixture to fill the gaps between the legs. The results revealed that at 18°C temperature difference, the generated output power of the developed TEG was $3.9 \mu\text{W}/\text{cm}^2$.



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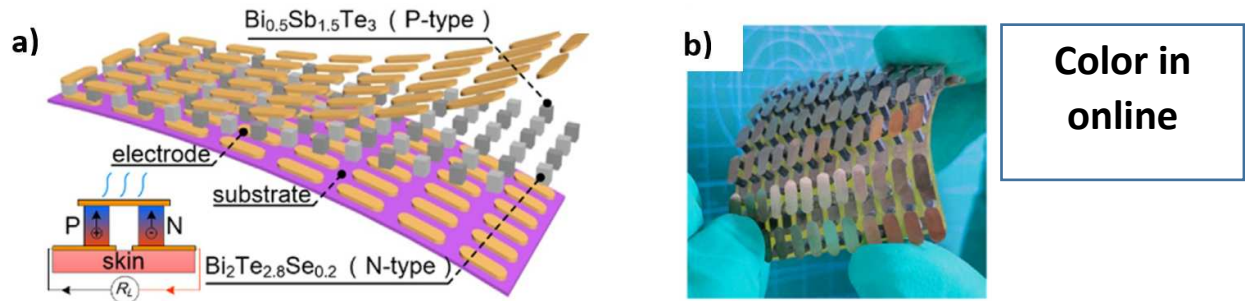
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Fig. 8. Combination of the PCB and the thermocouples [75].

304 Instead of encapsulating the thermocouples in a single PDMS block, Liu et al. [76] encapsulated each
 305 of them in an individual PDMS block to address the flexibility of the TEG. Accordingly, the authors
 306 soldered 18 pairs of ingot-shaped and Bi_2Te_3 -based legs each with the dimensions of $2\text{ mm} \times 2\text{ mm} \times$
 307 1.5 mm onto a FPCB. Then, the legs were connected together from their cold junctions with silver
 308 pastes as electrodes. To prevent electric shock, a polyimide film possessing super thermal and electrical
 309 resistivity was placed on the silver pastes. Finally, a thin (i.e. 1mm) layer of PDMS was poured around
 310 each thermocouple to encapsulate them in separate PDMS blocks. The results demonstrated that the
 311 as-prepared TEG generated the maximum power density of $3\ \mu\text{W}/\text{cm}^2$ at ΔT of 12°C .

312 To eliminate the negative impact of coating the legs with a polymer substrate, Yuan and Zhu [77]
 313 omitted the PDMS encapsulation and left the legs exposed to the ambient air, see Fig. 9. To illustrate,
 314 47 pairs of *p*-type ($\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$) and *n*-type ($\text{Bi}_2\text{Te}_{2.8}\text{Se}_{0.2}$) legs were soldered onto a FPCB from the
 315 bottom. Next, the legs were electrically connected together from the top by being soldered onto copper
 316 strips. Leaving the legs exposed to the ambient air, the results showed that the power density of the
 317 TEG at 14°C temperature gradient was $3.5\ \mu\text{W}/\text{cm}^2$. In addition, it was demonstrated that there was a
 318 direct relationship between the output voltage of the TEG and the legs' height. However, there was a
 319 converse relationship between its output voltage and fill factor. Precisely, when the fill factor was 4%,
 320 increasing the legs' height from 1 mm to 3mm raised the voltage by five times. Whereas, this increase
 321 was less than one half by 20% fill factor.

322



323

324

Fig. 9. a) Schematic view of the TEG's component; b) Photo of the fabricated TEG [77].

325

326 Shi et al. [61] applied the same principles to fabricate a flexible and wearable TEG. However, the
327 number of the thermocouples reduced to 24, and the utilized *p*-type and *n*-type TE materials were
328 $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ and $\text{Bi}_2\text{Se}_{0.5}\text{Te}_{2.5}$ composites, respectively. The results indicated that the fabricated TEG
329 achieved the highest output power of $4.75 \mu\text{W}/\text{cm}^2$ at ΔT of 35°C . Furthermore, the authors
330 demonstrated that by wearing the fabricated TEG on the human wrist, it generated 10.5mV that was
331 adequate for powering an LED.

332 Instead of holding the legs in place by a polymer-based substrate, Eom et al. [78] developed a flexible
333 holder in the shape of a flexible bracelet, see Fig. 10. The bracelet consisted of ten separate modular
334 units, and each unit held four ingot-shaped TE legs arranged in an array of 2×2 . It should be noted
335 that the *p*- and *n*-type legs were respectively composed of $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ and $\text{Bi}_2\text{Te}_{2.7}\text{Se}_{0.3}$ composites.
336 The legs were electrically connected together from the top and the bottom with copper foils.
337 Integrating the ten units (each with the surface area of $14 \text{ mm} \times 12 \text{ mm}$) together, a shaft was passed
338 through the punched holes in the legs and sides of the units. Finally, top of the legs was covered with
339 a copper sheet as the heatsink. The results showed that at ΔT of 20°C , the highest obtained output
340 powers under sitting, walking and running conditions were $2 \mu\text{W}/\text{cm}^2$, $2.9 \mu\text{W}/\text{cm}^2$, and $4.7 \mu\text{W}/\text{cm}^2$,
341 respectively.

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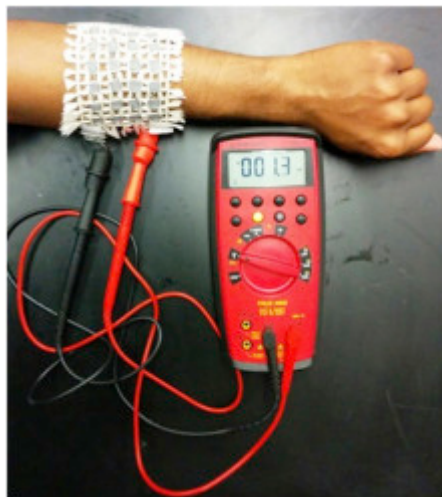
344

Fig. 10. The designed flexible TE module consisting a flexible holder [78].

345 Instead of preparing the ingot-shaped legs by cold/hot pressing of the TE powders, Siddique et al. [79]
346 prepared TE pastes and manually screen printed them into a pre-prepared fabric. To illustrate, the
347 authors demonstrated that they have used this technique due to its simplicity, low cost, and low curing
348 temperature (i.e. in the range of between 100-200°C). Accordingly, as Fig. 11 depicts, 24 holes were
349 cut in a thick polyester fabric to manually spread the *p*- and *n*-type pastes into them. Notably, the
350 employed TE materials were $(0.25\text{Bi},0.75\text{Sb})_2(0.95\text{Te},0.05\text{Se})_3$ for the *p*-type and $(0.98\text{Bi},0.02\text{Sb})_2$
351 $(0.9\text{Te},0.1\text{Se})_3$ for the *n*-type legs. After curing the pastes, the ingot-shaped legs were connected
352 together with a silver thread as the electrodes. The results showed that when the TEG was placed on
353 the arm, the highest achieved output power was $1 \times 10^{-5} \mu\text{W}/\text{cm}^2$ at ΔT of 32.9°C.

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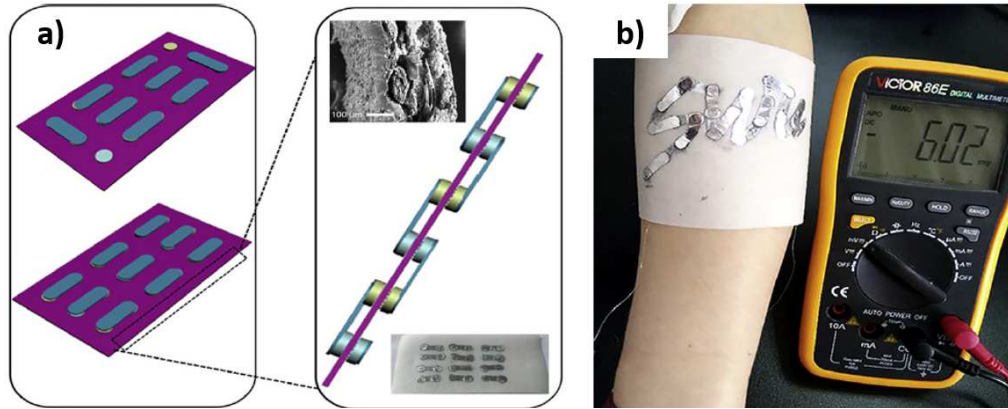


355

356

Fig. 11. The fabricated flexible TEG worn on the arm [79].

357 Likewise, Lu et al. [80] applied a manually dispenser printing technique to fill the 24 holes of a silk
 358 fabric with TE pastes. Accordingly, Bi_2Te_3 (*n*-type) and Sb_2Te_3 (*p*-type) pastes were prepared and
 359 deposited into the punched holes. Next, the legs were connected together with silver foils as
 360 interconnects, resulting in 12 thermocouples, see Fig. 12. The authors demonstrated that the fabricated
 361 TEG obtained the maximum output power of $4.7 \times 10^{-4} \mu\text{W}/\text{cm}^2$ at 35°C temperature gradient.



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Fig. 12. a) Schematic of the legs integration with the polyester fabric; b) Photo of the fabricated silk-based TEG [80].

365 3.1.1 Heatsink

366

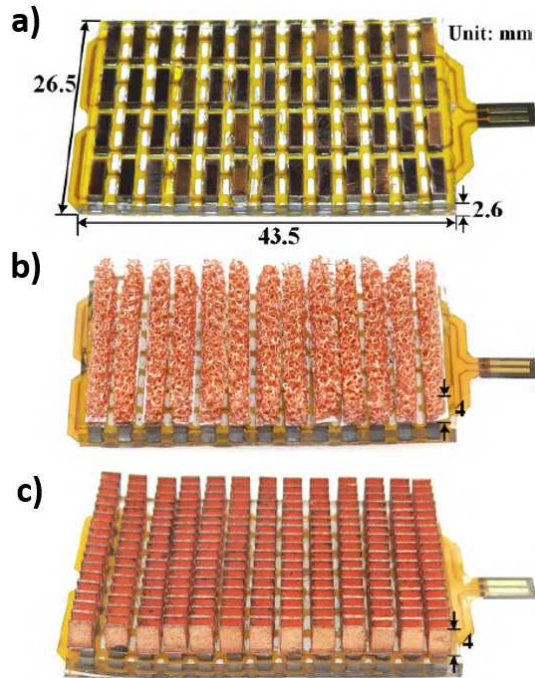
367 As mentioned before, a TEG generates a high open circuit voltage when a large temperature gradient
 368 exists between its hot and cold junctions. This high temperature difference could be obtained when a
 369 large heat flux is applied on the hot junctions, but at the same time the cold junctions dissipate a large
 370 amount of heat to the ambient. Accordingly, an effective heatsink is required to accelerate absorbing
 371 the thermal energy from the hotter side and releasing it to the cooler ambient. Notably, applying a
 372 heatsink with a low heat removal efficiency possibly damages the TE legs at high temperatures [81].
 373 Thus, in order to increase the rate of heat dissipation from heatsinks, it is necessary to increase either
 374 their thermal conductivity, heat transfer coefficient, or surface area [82]. Concerning wearable TEGs,
 375 the heatsinks are cooled either by phase change cooling (active cooling) or natural air cooling (passive
 376 cooling) techniques. Therefore, herein, it is intended to focus on the studies that designed heatsinks
 377 particularly to cool down wearable TEGs. It is noteworthy that this sub-section (i.e. 3.1) is selected for
 378 raising this topic because so far heatsinks have been only fabricated onto the wearable TEGs
 379 comprising inorganic ingot-shaped legs. Regarding the air cooled heatsinks, they are prepared in the

380 form of either a fin-shaped anodized aluminium, a high thermal conductive polymer-based substrate,
381 a copper sheet, a copper foam block, a copper plate-fins, or a cooling gel. For example, Nozariasbmarz
382 et al. [83] fabricated an aluminium plate and a fin-shaped anodized aluminium heatsink respectively
383 onto the cold substrate of a conventional rigid TEG. The TEG consisted of 6 pairs of Bi₂Te₃ -based
384 TE legs that were electrically connected together with gold-coated copper strips as electrodes. The
385 authors defined two fill factors for the investigation, including 36% and 12%. The tests were conducted
386 on different parts of the body at 17°C ambient temperature. The results showed that the 12% fill factor
387 was superior to the 36% one regarding the output power. Precisely, the former generated 34 μW/cm²,
388 16 μW/cm², 14 μW/cm², and 12 μW/cm² power densities when placed on the arm, foot, forearm, and
389 wrist, respectively. In addition, the 12% fill factor provided greater temperature gradient (i.e. ΔT =
390 2.5°C) along the legs than the 36% one (i.e. ΔT = 1.2°C).

391 However, the ongoing progress in flexible wearable TEGs has triggered extensive research on flexible
392 heatsinks. For instance, instead of using rigid aluminium-based heatsinks, Shi et al. [84] developed
393 flexible copper-based heatsinks comprising of either copper foam blocks or copper plate-fins, see Fig.
394 13. In fact, the authors developed these two heatsinks for the flexible cross-plane TEG that they had
395 developed in their previous research [75]. While the overall dimensions of the TEG were 26.5 mm ×
396 43.5 mm × 2.6 mm, the thickness of both heatsinks was approximately 4 mm. The heatsinks were
397 separately adhered to the top surface of the pre-developed TEG using a 0.1 mm thick thermal
398 conductive adhesive. The results showed that both heatsinks slightly improved the output power of the
399 TEG at temperature differences higher than 15°C. However, the foam block heatsink was slightly
400 superior to the plate-fins one, because it achieved the highest output power of 3.5 μW/cm² at ΔT of
401 20°C.

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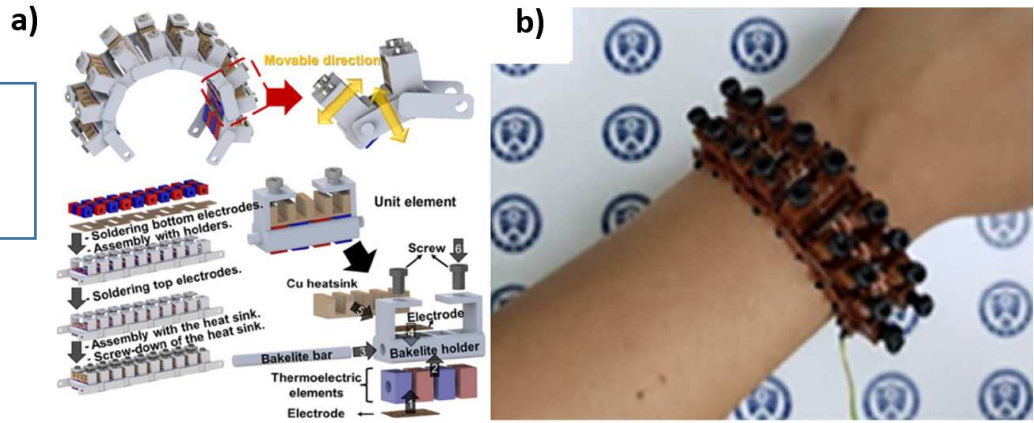


403
404 **Fig. 13.** Fabricated wearable TEGs: a) without heatsink; b) with copper-foam heatsink; and c) with plate-fins heatsink
405 [84].
406

407 Instead of applying a thermal conductive adhesive to couple the copper-based heatsink with the flexible
408 TEG, Park et al. [85] screwed the heatsink into the TEG. To clarify, 20 couples of ingot-shaped legs
409 were soldered onto a FPCB served as the bottom electrode. Then, the thermocouples were inserted into
410 the 40 holes of a bendable Bakelite holder arranged in an array of 4×10 . It should be noted that the
411 legs were composed of *p*-type $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ and *n*-type $\text{Bi}_2\text{Te}_{2.7}\text{Se}_{0.3}$ composites. Next, the legs were
412 electrically connected together from their top sides with copper films. Finally, a fin-shaped copper
413 heatsink was placed on the copper films and screwed into the Bakelite holder, as Fig. 14 depicts. The
414 authors demonstrated that the developed TEG with approximately $79 \text{ mm} \times 17 \text{ mm}$ base area obtained
415 the highest output power of $6.97 \mu\text{W}/\text{cm}^2$ at ΔT of 12.5°C .

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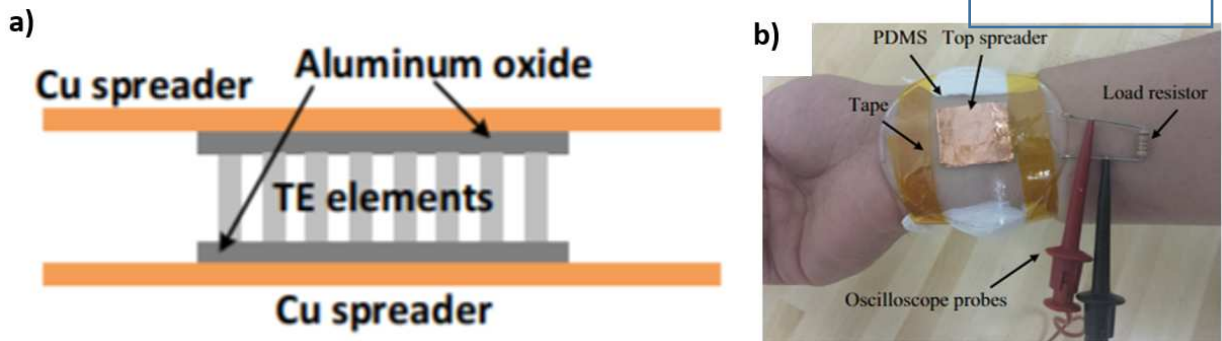


417
418 **Fig. 14.** a) Schematic of the entire device and its fabrication processes; b) The fabricated device worn on the wrist [85].
419

420 To increase the flexibility of the copper-based heatsinks, Hyland et al. [47] replaced the 3D copper
421 heatsinks with two copper sheets, see Fig. 15. In particular, the authors sandwiched 25 Bi₂Te₃-based
422 thermocouples between two rigid Aluminium Oxide ceramic substrates. Then, two separate copper
423 sheets were attached to the cold and hot substrates of the TEG as the heatsinks. The results proved that
424 the TEG generated higher output power compared with that of without a heatsink. Furthermore,
425 concerning the output power, there was a direct relationship between the heatsinks' size and the air
426 velocity. The paper concluded that at roughly 18°C temperature gradient, the developed TEG generated
427 the output powers of 6μW/cm², 5.5μW/cm², 5μW/cm², and 1.5 μW/cm² when placed on the wrist,
428 upper arm, chest, and T-shirt, respectively.

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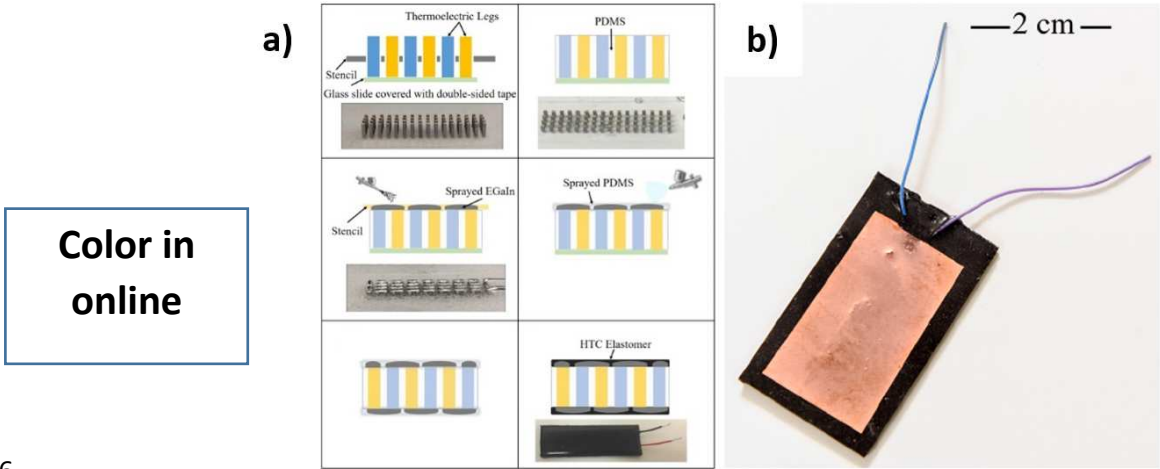
430
431 **Fig. 15.** a) Schematic of the developed TEG; b) The experimental setup of the TEG on the arm [47].
432

433 Likewise, Settaluri et al. [86] used two separate copper sheets to perform both as the heatsinks and the
434 integrating units. The TEG consisted of 256 Bi₂Te₃-based thermocouples that were electrically
435 connected together with copper strips and sandwiched between two ceramic substrates. Both substrates
436 were covered with separate copper plates (each with the dimensions of 292 mm × 964 mm) as the
437 heatsinks. Then, the gap between the two copper plates was filled with a flexible thermal insulator.
438 The authors determined three individual grooving patterns for the upper copper plate, namely; flat,
439 stripe, and checkerboard. The results showed that at 10°C temperature gradient, the highest achieved
440 output powers by the checkerboard, strip, and flat patterns were 28.5 μW/cm², 21.6 μW/cm², and 17.6
441 μW/cm², respectively.

442 As mentioned before, one of the most promising approach to flexibly hold the legs in place is applying
443 polymer-based substrates. Jeong et al. [87] used the same approach to develop a flexible polymer-
444 based heatsink. Precisely, the authors utilized a polymer-based substrate that simultaneously served as
445 a flexible heatsink and the coating layer of the electrodes. To specify, 8 pairs of Bi₂Te₃-based TE legs
446 were electrically connected together using Gallium-based liquid alloy. Then, the gaps between the
447 ingot-shaped legs were filled with the polymer-based plain Ecoflex substrate. However, the electrodes
448 were coated with EcoFlex: Gallium substrates, which possessed a higher thermal conductivity than the
449 plain Ecoflex substrate. As a result, due to the higher thermal conductivity of the outmost EcoFlex:
450 Gallium layers than the middle plain Ecoflex layer, the outmost layers performed as flexible heatsinks.
451 The results showed that the maximum output power of the fabricated TEG was 40.6 μW/cm² at ΔT of
452 20°C. Whereas, coating both the legs and the electrodes with the plain EcoFlex substrate resulted in
453 the remarkably lower output power of 19.8 μW/cm².

454 Likewise, Sargolzaeiaval et al. [88] encapsulated the electrodes in a flexible high thermal conductivity
455 mixture to serve both as a coating substrate and a flexible heatsink. To specify, the ingot-shaped *p*- and
456 *n*-type TE legs were composed of Bi_{0.5}Sb_{1.5}Te₃ and Bi₂Se_{0.3}Te_{2.7} composites, respectively. After
457 arranging the 32 thermocouples in an array of 4×16, they were connected together with EGaIn as liquid
458 metal electrodes. Then, the spaces between them were filled by PDMS, leaving the electrodes exposed
459 to the ambient air. To examine the impact of the coating substrate of the electrodes on the TE
460 performance of the TEG, two individual substrates were determined to coat the EGaIn electrodes,
461 including plain PDMS and PDMS: EGaIn mixture. Concerning the output power of the TEG, the
462 results showed the superiority of the latter to the former. This was mainly because the latter possessed

463 higher thermal conductivity than the former. The authors also fabricated a thin copper sheet onto the
 464 top PDMS: EGeIn layer to intensify the heat dissipation from the cold junctions, see Fig. 16. The
 465 results illustrated that $5 \mu\text{W}/\text{cm}^2$ was the highest output power achieved by the TEG at ΔT of 11°C .



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 467 **Fig. 16.** a) Fabrication processes of the TEG; b) Photo of the fabricated TEG with copper sheet as the heatsink [88].

468
 469 Increasing the thermal conductivity of the PDMS substrate, Lee et al. [89] replaced EGeIn with silver-
 470 nickel (Ag–Ni) particles. To specify, as soft heat conductors, Ag–Ni particles were initially added to
 471 the PDMS mixture followed by pouring the mixture into a pre-prepared mould. Then, silver-nanowires
 472 (Ag–NW) were mask printed as soft electrodes onto the mixture. After that, the Ag–Ni particles in the
 473 mixture were magnetically aligned in the direction of the legs to facilitate the heat transfer from the
 474 skin to the legs, as Fig. 17 illustrates. Next, using a fully automated epoxy printing technique, 440
 475 ingot-shaped and Bi_2Te_3 -based legs were integrated onto the Ag–NW electrodes followed by filling
 476 the gaps between them with plain PDMS. Then, silver-nanowires (Ag–NW) were mask printed onto
 477 the plain PDMS substrate as the top electrodes followed by pouring a layer of Ag–Ni: PDMS mixture
 478 onto it. Next, the Ag–Ni particles in the upper substrate were magnetically aligned in the direction of
 479 the legs to facilitate the heat transfer from the legs to the ambient. Thus, both the top and the bottom
 480 Ag–Ni: PDMS layers performed as flexible heatsinks. The results revealed that when the prepared
 481 flexible TEG with an area of $39 \times 43 \text{ mm}^2$ was mounted on the hand, it generated the highest power
 482 density of $6.96 \mu\text{W}/\text{cm}^2$ at 10°C temperature gradient.

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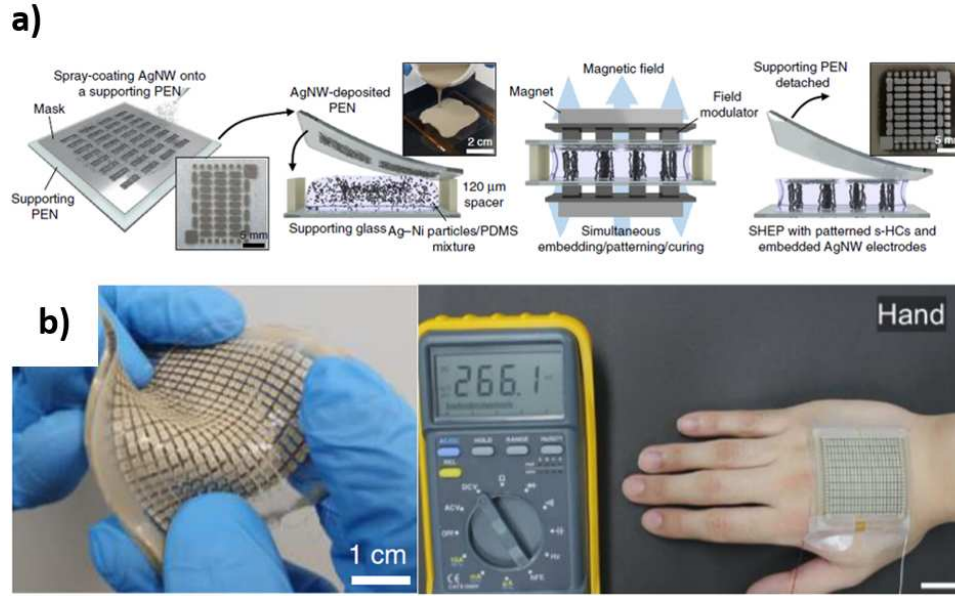


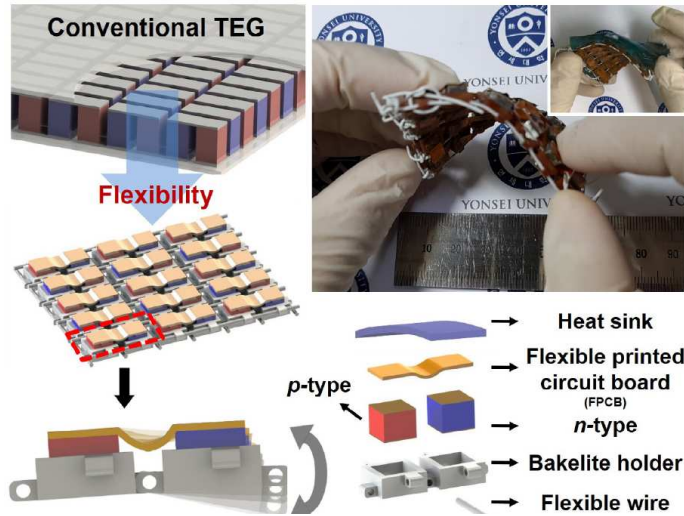
Fig. 17. a) Schematic illustration; and b) the photos of the fabricated TEG [89].

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However, rather than conductive cooling, Park et al. [90] applied evaporative cooling to cool down their fabricated wearable TEG. Notably, the authors used a water-absorbing polymer (i.e. hydrogel) as the heatsink to cool down the TEG based on liquid evaporation. Regarding hydrogels, they are considered as porous materials in which a fluid (e.g. water) is entrapped into a polymeric matrix [91]. In fact, hydrogels can absorb 10 to 1000 times its own weight in water [92]. In particular, the authors used *p*-type ($\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$) and *n*-type ($\text{Bi}_2\text{Te}_{2.7}\text{Se}_{0.3}$) ingot-shaped TE legs to fabricate a mat-shaped flexible TEG. The TEG consisted of 15 thermocouples that were placed inside Bakelite holders and electrically connected together using FPCBs, see Fig. 18. The holders were connected together by passing flexible wires through the punched holes in the opposite sides of the holders. Then, a layer of hydrogel was spread on the cold side of the TEG (i.e. over the upper FPCB) as the heatsink. Since the performance of hydrogel is based on liquid evaporation, the evaporation of the trapped water over time allowed the absorbed heat to be continuously released to the ambient air. Finally, the mat-shaped flexible TEG with $55 \text{ mm} \times 45 \text{ mm}$ base area was embedded in an armband to be mounted on the lower arm. The results showed that the highest output power of the TEG at 9.3°C temperature difference was $5.60 \mu\text{W}/\text{cm}^2$.

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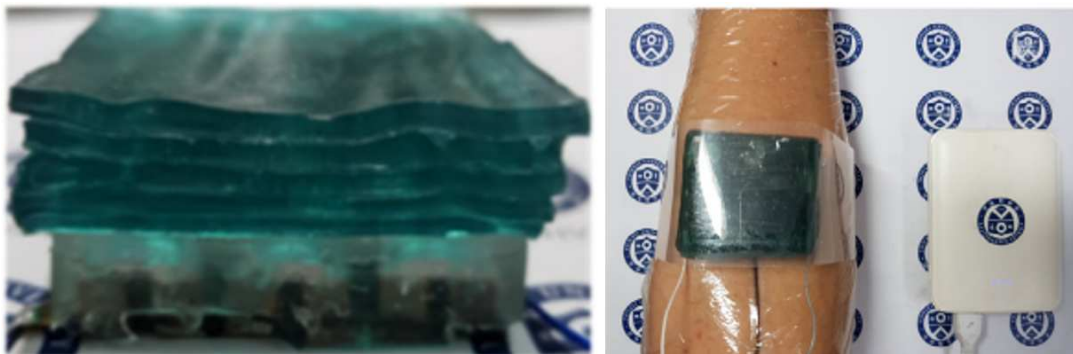
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Fig. 18. The consisting elements of the fabricated TEG and the photo of the developed prototype [90].

505 Likewise, Lee et al. [93] used hydrogel as the heatsink of their developed flexible wearable TEG.
506 Precisely, 15 pairs of ingot-shaped p - ($\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$) and n - ($\text{Bi}_2\text{Te}_{2.7}\text{Se}_{0.3}$) type legs were soldered onto
507 a FPCB as the bottom electrode. From the top, the legs were electrically connected together with
508 EGaIn. To hold the bulk legs in place, a flexible resin holder was placed around them followed by
509 filling the gaps between them with PDMS. While the PDMS substrate was coated the EGaIn
510 electrodes, a flexible hydrogel-based heatsink was fabricated onto it, see Fig. 19. When the TEG was
511 placed on the upper arm, the results demonstrated that it generated $8.30 \mu\text{W}/\text{cm}^2$ at 8°C temperature
512 difference.

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Fig. 19. Fabricated hydrogel-based heatsink onto the TEG [93].

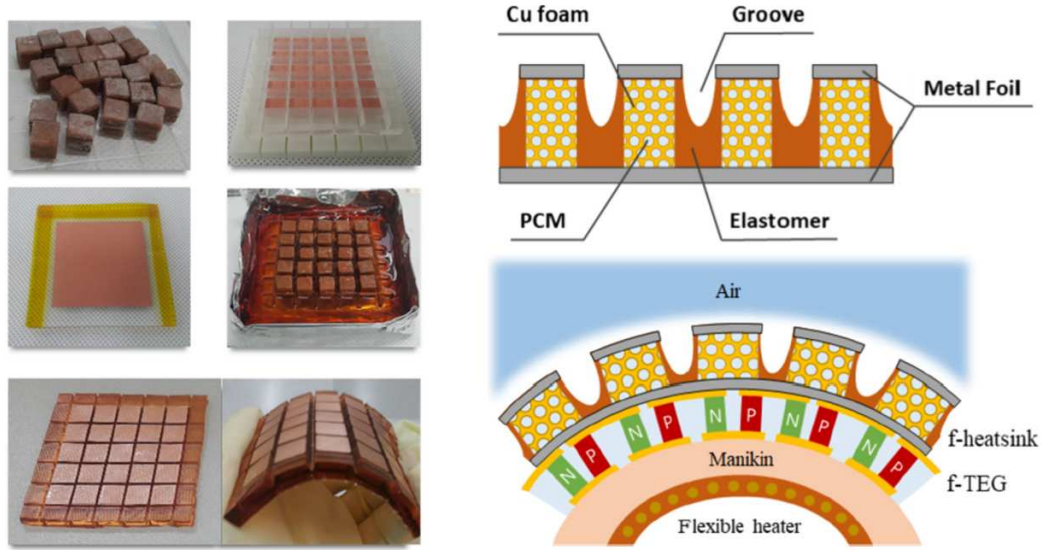
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517 Rather than spreading hydrogel on the cold side of the TEG, Kim et al. [94] embedded it within a fabric
518 to be coupled with the TEG. To illustrate, 170 ingot-shaped thermocouples were prepared from *p*-type
519 Bi_{0.5}Sb_{1.5}Te₃ and *n*-type Bi₂Te_{2.7}Se_{0.3} composites. While the thermocouples were connected together
520 with copper electrodes, the gaps between them were filled with a polymer-based material as the flexible
521 integrating substrate. Regarding the heatsink, the authors encapsulated a hydrogel material (e.g. a
522 superabsorbent polymer) inside a fabric followed by integrating it with the TEG from its cold side.
523 The results demonstrated that at 12°C temperature difference, the TEG generated over 38 μW/cm² and
524 13 μW/cm² after 10 min and over 22 hrs, respectively.

525 Instead of evaporative cooling, Lee et al. [95] used exothermic and endothermic reactions to maintain
526 a constant temperature on the cold side of the TEG over a period of time. Accordingly, the authors
527 used a phase change material (PCM) called n-octadecane (C₁₈H₃₈) as the heatsink to absorb latent
528 heat from the cold side of the TEG and release it to the ambient air. N-octadecane was selected as the
529 PCM due to its low melting point (28°C), resulting in a large amount of heat absorption at the low
530 temperature difference between the skin and ambient air. To fabricate the heatsink, 25 copper foam
531 blocks each with the dimensions of 10 mm × 10 mm × 5 mm were filled with the PCM. This was
532 because, the foam blocks could spread the heat uniformly within the PCM. Then, the filled foam blocks
533 were arrayed in an array of 5×5, and the spaces between them were filled with a polyurethane
534 elastomer. Next, the top and the bottom sides of the filled foam blocks were covered with a metal foil
535 to intensify the vertical heat flow, see Fig. 20. Finally, the generated heatsink was attached to the top
536 substrate of a cross-plane TEG comprising 50 ingot-shaped thermocouples. To specify, the legs were
537 Bi₂Te₃ -based and the gaps between them were filled with a polymer-based filler. The results showed
538 that combining the as-prepared heatsink with the 65 mm × 65 mm base area TEG resulted in an output
539 power of 20 μW/cm² at ΔT of 3°C.

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541
542 **Fig. 20.** Fabrication processes of the flexible heatsink consisting of copper foam blocks and PCM [95].
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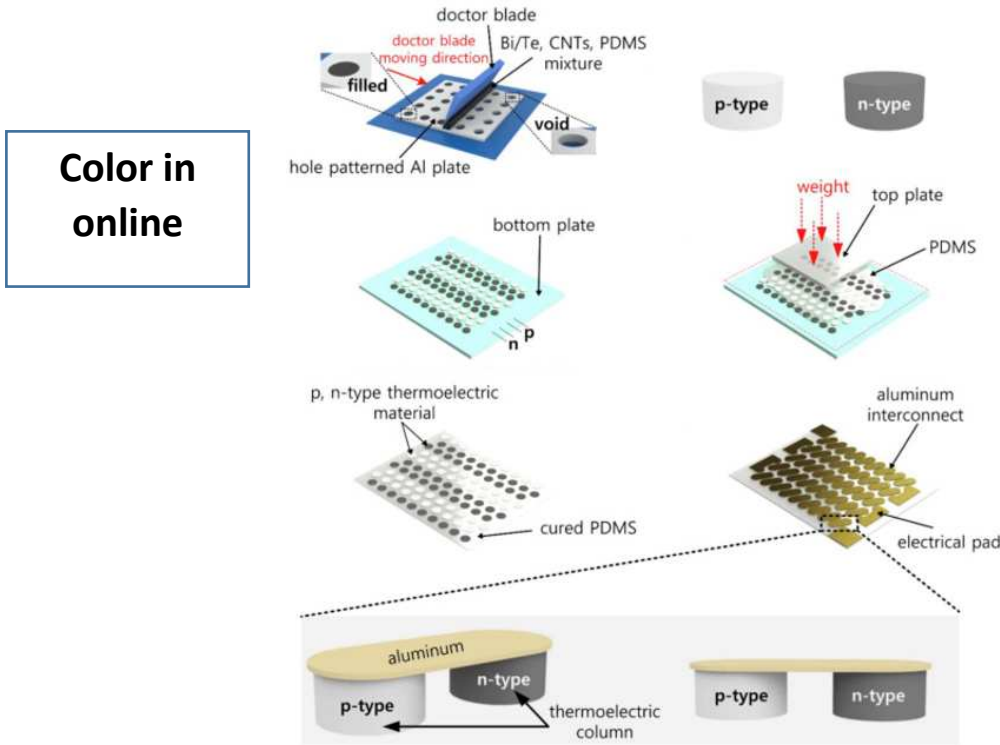
544 3.2. Organic TE materials

545 Although organic TE materials benefit from an intrinsic low thermal conductivity, it is not prevalent
546 to use them for the preparation of the ingot-shaped TE legs. This is mainly because their low thermal
547 conductivity could be replaced by the high power factor of the inorganic type. In addition, it will be
548 explained in section 4.2 that organic TE materials (e.g. polymers) possess a great mechanical
549 flexibility, thus it is too tough to overcome this feature to turn them into ingot-shaped TE legs.
550 Therefore, so far, no study has used a pristine organic TE material to prepare an ingot-shaped TE leg.

552 3.3. Hybrid TE materials

553 As reviewed in section 3.1, the inorganic ingot-shaped TE legs should be embedded in a flexible
554 substrate to address the flexibility requirement of wearable TEGs. However, when the TEG bends, a
555 bending force is applied on the flexible substrate due to the rigidity of the inorganic TE materials.
556 Precisely, the bending force is concentrated on the junctions of the rigid legs and the flexible substrate,
557 reducing the flexibility of the device. Allowing the bending force to distribute uniformly throughout
558 the TEG, Jung et al. [60] attempted to increase the flexibility of the ingot-shaped legs themselves by
559 applying hybrid TE materials. To specify, the authors replaced the inorganic ingot-shaped legs with
560 the hybrid ones to benefit both from the high conversion efficiency and the flexibility of the inorganic
561 and organic types, respectively. To prepare the hybrid TE pastes, the authors separately added Bi (*p*-
562 type) and Te (*n*-type) powders to CNTs followed by mixing them with PDMS. Then, the as-prepared

563 *p*- and *n*-type pastes were poured into 100 holes of an aluminium stencil followed by being cured for
 564 3 hrs to become stiff. Then, the stiffed legs were electrically connected together in series with
 565 aluminium strips, resulting in 50 thermocouples. Finally, the gaps between the legs were filled with
 566 PDMS as the flexible substrate, see Fig. 21. The results showed that the highest output power generated
 567 by the developed TEG was $80 \mu\text{W}/\text{cm}^2$ at ΔT of 293°C .



568
 569 **Fig. 21.** Schematic of the fabrication processes of the proposed TEG [60].
 570

571 Instead of preparing a hybrid TE material, Park et al. [96] connected 36 pairs of organic *p*-type and
 572 inorganic *n*-type legs together. In particular, the *p*-type legs were film-shaped and consisted of doped
 573 poly(3,4-ethylenedioxythiophene) (PEDOT) in tosylate. After preparing the *p*-type polymer film, it
 574 was cut into smaller area films ($18 \times 2 \text{ mm}^2$) to be individually rolled up in an elliptical shape. The *n*-
 575 type legs were composed of Bi_2Te_3 ingots with the dimensions of $2 \text{ mm} \times 2 \text{ mm} \times 0.2 \text{ mm}$. In accordance
 576 with the dimensions of the *n*-type legs, the polymer films were rolled up with a 4:1 ratio in the
 577 horizontal diameter (8 mm) against the vertical diameter (2 mm). The authors selected a polyimide
 578 film as a flexible substrate and coated it with a gold sheet in a specific pattern. To electrically connect
 579 the 36 prepared thermocouples together, they were soldered onto a gold sheet from the bottom and
 580 wired with a polytetrafluoroethylene coated wire from the top. The results showed that when the TEG

581 was worn on the wrist, it generated an output voltage of 10.6 mV. [Table 2](#) illustrates the summary of
582 the reviewed literatures on wearable TEGs consisting ingot-shaped legs.

583

584

585 **Table 2.** Summary of the recently developed wearable TEGs comprising ingot-shaped thermocouples.

586

<i>P</i> -type	<i>N</i> -type	Couple Number	Electrode	Packaging	Heatsink	ΔT (°C)	Output voltage or power	Ref
<i>Inorganic</i>								
Bi ₂ Te ₃	Bi ₂ Te ₃	25	Copper	Rigid ceramic substrates	Copper sheet	18	6 $\mu\text{W}/\text{cm}^2$	[47]
Bi _{0.5} Sb _{1.5} Te ₃	Bi ₂ Se _{0.5} Te _{2.5}	24	Copper	FPCB	n/a	35	4.75 $\mu\text{W}/\text{cm}^2$	[61]
Bi _{0.3} Sb _{1.7} Te ₃	Bi ₂ Sb _{0.3} Te _{2.7}	72	Copper	Flexible polymer	n/a	8.9	2.5 $\mu\text{W}/\text{cm}^2$	[71]
Bi _{0.5} Sb _{1.5} Te ₃	Bi ₂ Se _{0.3} Te _{2.7}	32	EGaIn	PDMS	n/a	13	1 $\mu\text{W}/\text{cm}^2$	[72]
Bi ₂ Te ₃	Bi ₂ Te ₃	47	Copper	PDMS sponge	n/a	8	130 $\mu\text{W}/\text{cm}^2$	[73]
Bi _{0.5} Sb _{1.5} Te ₃	Bi ₂ Te _{2.7} Se _{0.3}	24	Copper	Flexible polymer	n/a	10	4.5 $\mu\text{W}/\text{cm}^2$	[74]
Bi _{0.5} Sb _{1.5} Te ₃	Bi ₂ Se _{0.5} Te _{2.5}	52	Copper	FPCB + PDMS	n/a	18	3.9 $\mu\text{W}/\text{cm}^2$	[75]
Bi ₂ Te ₃	Bi ₂ Te ₃	18	Silver paste and Copper	FPCB + PDMS	n/a	12	3 $\mu\text{W}/\text{cm}^2$	[76]
Bi _{0.5} Sb _{1.5} Te ₃	Bi ₂ Te _{2.8} Se _{0.2}	47	Copper	FPCB	n/a	14	3.5 $\mu\text{W}/\text{cm}^2$	[77]
Bi _{0.5} Sb _{1.5} Te ₃	Bi ₂ Te _{2.7} Se _{0.3}	20	Copper	Flexible holder (Bakelite)	n/a	20	2 $\mu\text{W}/\text{cm}^2$	[78]
(0.25Bi,0.75Sb) ₂ (0.95Te,0.05Se) ₃	(0.98Bi,0.02Sb) ₂ (0.9Te,0.1Se) ₃	12	Silver thread	Polyester fabric	n/a	32.9	1*10 ⁻⁵ $\mu\text{W}/\text{cm}^2$	[79]
Sb ₂ Te ₃	Bi ₂ Te ₃	12	Silver foils	Silk fabric	n/a	35	4.7×10 ⁻⁴ $\mu\text{W}/\text{cm}^2$	[80]
Bi ₂ Te ₃	Bi ₂ Te ₃	6	Gold-coated copper	Rigid ceramic substrates	Fin-type anodized aluminium	2.5	34 $\mu\text{W}/\text{cm}^2$	[83]
Bi _{0.5} Sb _{1.5} Te ₃	Bi ₂ Se _{0.5} Te _{2.5}	52	Copper	FPCB + PDMS	Copper foam	3.5	20 $\mu\text{W}/\text{cm}^2$	[84]
Bi _{0.5} Sb _{1.5} Te ₃	Bi ₂ Te _{2.7} Se _{0.3}	20	Copper	Flexible holder (Bakelite)	Fin-type copper	12.5	6.97 $\mu\text{W}/\text{cm}^2$	[85]
Bi ₂ Te ₃	Bi ₂ Te ₃	256	Copper	Rigid ceramic substrates and Thermal Insulator	Copper sheet	10	28.5 $\mu\text{W}/\text{cm}^2$	[86]
Bi ₂ Te ₃	Bi ₂ Te ₃	8	Gallium-based liquid alloy	Flexible polymer	EcoFlex: Gallium Layer	20	40.6 $\mu\text{W}/\text{cm}^2$	[87]
Bi _{0.5} Sb _{1.5} Te ₃	Bi ₂ Se _{0.3} Te _{2.7}	32	EGaIn	PDMS	PDMS: EGaIn Layer and Copper Sheet	11	5 $\mu\text{W}/\text{cm}^2$	[88]
Bi ₂ Te ₃	Bi ₂ Te ₃	220	Silver-nanowires	PDMS	Ag–Ni :PDMS Layer	10	6.96 $\mu\text{W}/\text{cm}^2$	[89]

$\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$	$\text{Bi}_2\text{Te}_{2.7}\text{Se}_{0.3}$	15	Soldered to FPCB	Flexible holder (Bakelite)	Solid-state cool gel	9.3	5.6 $\mu\text{W}/\text{cm}^2$	[90]
$\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$	$\text{Bi}_2\text{Te}_{2.7}\text{Se}_{0.3}$	15	Soldered to FPCB from bottom and connected with EGaIn from top	PDMS and resin holder	Hydrogel	8	8.30 $\mu\text{W}/\text{cm}^2$	[93]
$\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$	$\text{Bi}_2\text{Te}_{2.7}\text{Se}_{0.3}$	170	Copper	Flexible polymer	Liquid evaporative polymer	12	13 $\mu\text{W}/\text{cm}^2$	[94]
Bi_2Te_3	Bi_2Te_3	50	Copper	Flexible polymer	Phase change material	3	20 $\mu\text{W}/\text{cm}^2$	[95]
Hybrid								
Bi: CNTs: PDMS	Te :CNTs: PDMS	50	Aluminium	PDMS	n/a	293	80 $\mu\text{W}/\text{cm}^2$	[60]
PEDOT doped in tosylate	Bi_2Te_3	36	Gold and Flexible wire	Polyimide film	n/a	36	10.6 mV	[96]

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589 4. Film-shaped thermocouples

590

591 4.1. Inorganic TE materials

592

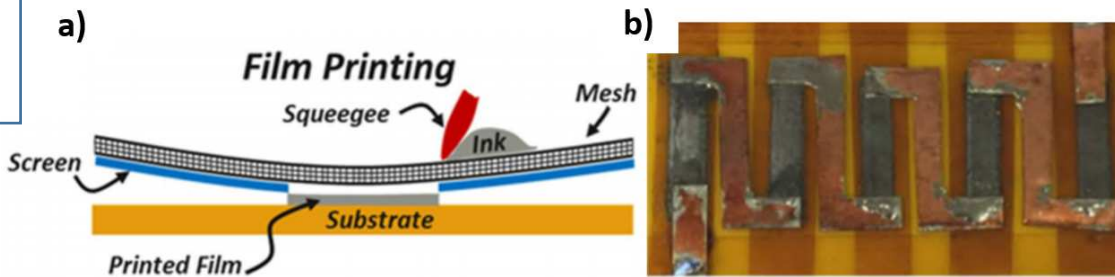
593 As mentioned before, flexible wearable TEGs have many advantages over the rigid ones due to their
594 flexibility, allowing them to conform to the arbitrary surfaces of the body. Accordingly, as reviewed
595 in Section 3, a flexible wearable TEG can be fabricated by embedding ingot-shaped thermocouples in
596 a flexible polymer-based substrate such as PDMS. To further increase the flexibility of wearable TEGs
597 and reduce the bending force, the ingot-shaped thermocouples can be replaced by the film-shaped ones.
598 Accordingly, various printing techniques can be applied to deposit TE materials onto flexible
599 substrates (e.g. plastic films or fabrics). These printing techniques are namely, dispenser printing,
600 inkjet printing, screen printing, roll to roll printing, and aerosol jet printing [97]. All these printing
601 techniques aim to synthesize high-performance TE inks and obtain printed TE films with high density
602 and electrical conductivity [98].

603 Regarding the screen printing technique, TE inks are pushed through the openings of a stencil or a
604 patterned mesh onto a flexible substrate. Precisely, a fill blade or squeegee is used to force the viscous
605 ink through the mesh, onto the substrate. The advantages of screen printing, as compared to the other

606 printing methods, are its compatibility with a broader range of available substrates and inks as well as
607 allowing greater ink thickness [99]. Notably, film-shaped TEGs are fabricated either unipolar or
608 bipolar, depending on the applied TE material.

609 Accordingly, Varghese et al. [98] developed a unipolar film-shaped TEG by applying the screen
610 printing technique. Precisely, the authors employed microwave stimulated wet chemical method to
611 prepare *n*-type Bi₂Te_{2.8}Se_{0.2} nanocrystals. Then, the prepared *n*-type ink was screen printed onto a
612 flexible polyimide substrate, generating 5 film-based TE legs each with the dimensions of 10 mm × 2
613 mm × 0.01 mm, see Fig. 22. To remove any solvent and binders, the printed TE films were dried on
614 a hot plate at 200°C. Next, the films were consolidated by cold compaction followed by being sintered
615 in the vacuum at 430°C for 45 min. To electrically connect the TE films together, the authors soldered
616 them to thin copper foils as electrodes. The results revealed that at a temperature difference of 20°C,
617 500 μW/cm² was the greatest achieved output power of the TEG.

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Fig. 22. a) Schematic of the screen printing process; and b) photo of the fabricated TEG [98].

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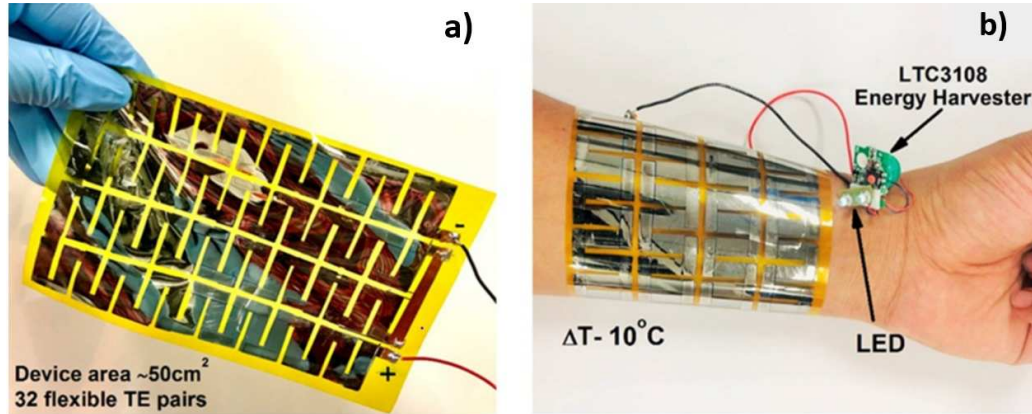
621 Guo et al. [100] replaced the *n*-type TE material with a *p*-type one called molybdenum disulphide
622 (MoS₂) to fabricate it onto a fibre-based underlying substrate. To illustrate, the authors added
623 H₂AuCl₄·4H₂O to MoS₂ to improve its TE performance, resulting in an Au decorated 2D MoS₂ nano-
624 sheets. Next, the Au-MoS₂ solution was deposited onto a 2D support unit via vacuum filtration to form
625 the TE films. The authors then cut five strips (5 mm × 45 mm) out of the *p*-type Au-MoS₂ film and
626 crossed them into the five pierced holes (6 mm × 1 mm) in a fabric. Then, a copper adhesive tape was
627 used to attach the two ends of the Au-MoS₂ strips to the sides of the fabric. Finally, the *p*-type strips
628 were electrically connected together with a commercial metal yarn. The results proved that the
629 fabricated TEG comprising five TE legs achieved the maximum output voltage of 2.5mV at ΔT of 5°C.

630 Instead of a unipolar film-shaped TEG, Cao et al. [101] developed a bipolar one by alternately screen
631 printing $\text{Bi}_{1.8}\text{Te}_{3.2}$ (*n*-type) and Sb_2Te_3 (*p*-type) pastes onto a polyimide substrate. To specify, the TE
632 pastes were alternately poured over the stencil and forced into its openings by a blade. To smooth the
633 outmost surface of the pastes and evenly squeeze them into the openings, a pressure squeegee passed
634 across the stencil. After lifting off the stencil, the experiment was finished by drying the pasted
635 substrate at 80°C for 2 min followed by curing it in an N_2 atmosphere at 253°C for 3 hrs. Finally, the
636 cured pastes were connected together with SbTe electrodes to generate 8 film-shaped thermocouples.
637 The results illustrated that at 20°C temperature gradient, $0.016 \mu\text{W}/\text{cm}^2$ was the maximum achieved
638 output power of the TEG.

639 Concerning the inkjet printing technique, it is a derivative of the dispenser printing technique where
640 ink drops are transferred onto a substrate by a nozzle. Adjusting expansion/contraction of the
641 piezoelectric actuator, a single ink droplet can be ejected from the nozzle forming the desired pattern
642 on the substrate [102]. Thus, Chen et al. [103] applied the inkjet printing technique to transfer Bi_2Te_3
643 (*n*-type) and $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ (*p*-type) nanowire inks onto a polyimide substrate. To improve the TE
644 properties of the 5 film-shaped thermocouples, they were annealed in a tube furnace using forming
645 gas. Notably, EGaIn liquid metal was used as electrodes to connect the films together. Finally, the
646 developed TEG was coated by a silicon elastomer to be protected from the ambient conditions. The
647 results showed that at 7°C temperature difference, the printed TEG delivered the maximum output
648 power of 14.1 nW.

649 Thermal evaporation is another technique to deposit TE thin films onto flexible substrates. In this
650 technique, the TE material is evaporated in a vacuum, where allows the vapour particles to travel to
651 the target substrate and condense back to the solid state in the form of a thin film [104]. Therefore,
652 Karthikeyan et al. [105] applied this technique to deposit *p*-type Tin telluride(SnTe) and *n*-type Lead
653 Telluride (PbTe) thin films onto a flexible polyimide substrate. Next, the 64 prepared legs were
654 connected together with aluminium films as electrodes, see Fig. 23. The results showed that the
655 fabricated TEG delivered $8.5 \text{ mW}/\text{cm}^2$ at a temperature difference of 120°C .

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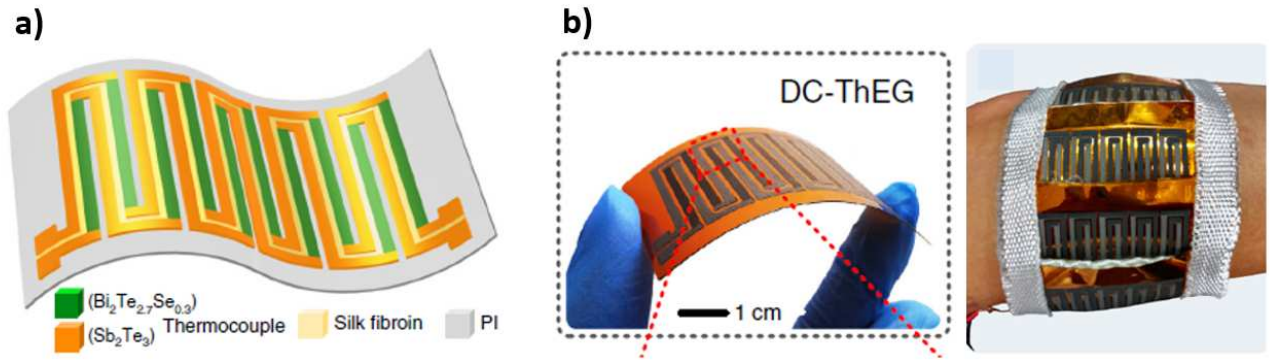
657
658 **Fig. 23.** a) Photo of the fabricated flexible SnTe–PbTe based TEG; b) Photo of the TEG placed on the arm as the power
659 source of an LED [105].

660
661 Likewise, Ren et al. [106] used thermal evaporation to fabricate 14 TE chips onto a polyimide film.
662 Precisely, each TE chip was generated by alternately thermal evaporating 4 pairs of $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ (*p*-
663 type) and $\text{Bi}_2\text{Te}_{2.8}\text{Se}_{0.3}$ (*n*-type) thin films onto the polyimide film. Then, the TE legs of each chip
664 were connected together with Au-Ge electrodes. To integrate the 14 TE chips together, 14 slots were
665 laser cut in the polyimide substrate. Next, the opposite ends of the alternate slots were connected
666 together by screen-printing liquid metal as electrodes. After that, the chips were slightly inserted into
667 the slots and stiffly bonded with the polyimide film by dropping a small amount of the polyimide
668 solution at the junctions. The results proved that at 93°C temperature gradient, the fabricated TEG
669 with 56 thermocouples generated the highest output power of $18.625 \mu\text{W}/\text{cm}^2$.

670 So far, all of the reviewed wearable TEGs only served as power sources of a sensor network. However,
671 none of them served as a sensor themselves along with being a power source. To fill this gap, Wen et
672 al. [107] replaced the single-chain thermocouples of film-shaped TEGs with complementary double-
673 chain thermocouples to provide a space for fabricating a dielectric material as a temperature &
674 humidity sensor. To specify, the authors screen printed *n*-type $\text{Bi}_2\text{Te}_{2.7}\text{Se}_{0.3}$ and *p*-type Sb_2Te_3 pastes
675 onto a polyimide film. In particular, the *n*- and *p*-type TE pastes were alternately screen printed onto
676 the underlying substrate in two separate complementary chains, see Fig. 24. While each individual TE
677 chain possessed 5 thermocouples, the gap between the two chains was filled by silk fibroin solution.
678 Notably, silk fibroin served as the dielectric material, changing (i.e. increasing/decreasing) at different
679 air temperatures and humidity. The results demonstrated that the developed wearable TEG with one
680 complementary double-chain thermocouple achieved the highest output power of $0.095 \mu\text{W}/\text{cm}^2$ at ΔT

681 of 20°C. Regarding the silk fibroin, absorbing and desorbing water molecules intensified and lessened
682 its molecular motion, leading to an increase and a decrease in its dielectric constant, respectively.
683 Likewise, increasing and decreasing the air temperature raised and reduced the dielectric constant of
684 the silk fibroin, resulting in an increase and a decrease in the capacitance of the double-chain TEG,
685 respectively.

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689 **Fig. 24.** a) Schematic illustration; and b) the photos of the fabricated TEG [107].

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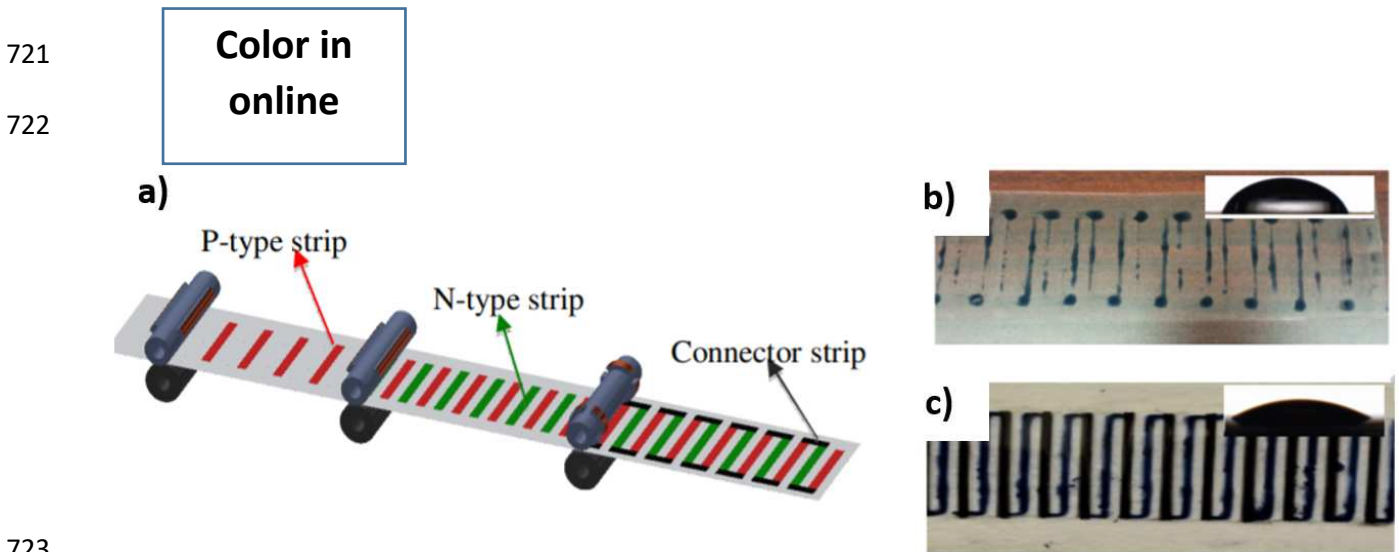
692 4.2. Organic TE materials

693
694 Although inorganic TE materials possess high energy conversion efficiency, they are intrinsically rigid
695 and unsuitable for enduring a wide range of dynamic motions. This is mainly attributed to their limited
696 elasticity and tensile strength as well as great hardness [108]. As a result, they cause a considerable
697 bending force at their junctions with flexible substrates, resulting in lower flexibility of TE modules
698 or cracks around the junctions. To overcome these issues, many researchers attempted to further
699 improve the flexibility of the film-shaped legs via preparing them with flexible TE materials (i.e.
700 organic TE materials). To specify, organic TE materials (e.g. conducting and non-conducting
701 polymers) benefit from unique merits such as inexpensive and convenient manufacturing process,
702 lightweight, mechanical flexibility, and intrinsically low thermal conductivity [109].

703 Accordingly, Elmoughni et al. [110] used polymer-based TE materials to fabricate 16 film-shaped
704 thermocouples onto a polyester knitted fabric. For the *p*-type films, dry pellets (8 wt%) of poly(3,4-

705 ethylenedioxythiophene) polystyrene sulfonate (PEDOT: PSS) were added to 5 vol% DMSO followed
706 by mixing the dispersion for 15 min in a micro vibration mill. For the *n*-type films, Poly[Na(NiETT)]
707 ink was synthesized, while its viscosity was optimized by micro ball milling of the ETT in
708 PVDF/DMSO solution at a weight ratio of 4:1 (ETT:PVDF). To fabricate the TEG, initially 32 holes
709 were burnt out of a polyester fabric to deposit the *p*- and *n*-type inks into them. Then, the *p*- and *n*-type
710 inks were alternately deposited into the holes using the screen printing technique. This step was
711 repeated on the other side of the fabric to ensure the penetration of the ink into the holes. Finally, the
712 TE legs were electrically connected together by hot pressing silicone-based silver ink at the ends of
713 the legs. The results revealed that the developed prototype yielded 3mV at ΔT of 3°C.

714 As a derivative of the screen printing technique, Zhang et al. [111] used roll-to-roll printing to print
715 PEDOT: PSS (*p*-type) and nitrogen-doped graphene (*n*-type) inks onto a flexible plastic substrate, see
716 Fig. 25. Concerning roll to roll printing, a rotary screen is used to transfer the ink continuously to the
717 substrate with tuneable rotation pressure and speed. Then, the TE legs were electrically connected
718 together using PEDOT: PSS aqueous solutions. After that, the substrate was treated alternately by UV
719 irradiation and oxygen plasma treatment for 2 hrs in total. The authors reported that the 18 printed
720 thermocouples obtained the maximum output power of $\sim 0.024 \mu\text{W}/\text{cm}^2$ at ΔT of 10°C.

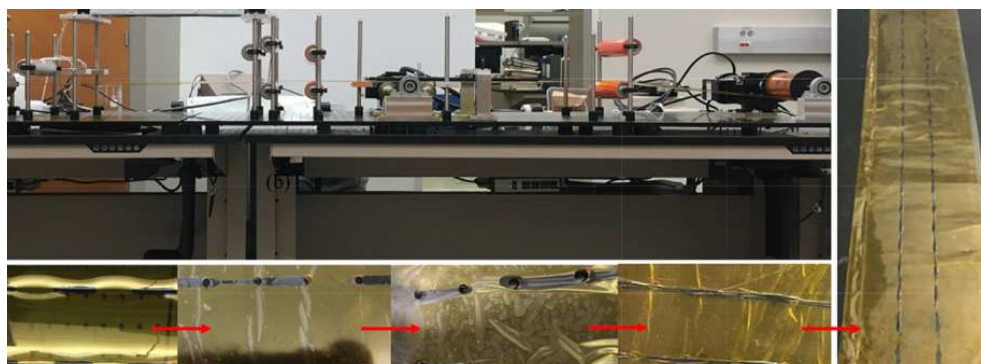


723
724 **Fig. 25.** a) Illustration of roll to roll printing of the flexible TE devices; b) UV treated plastic substrate; c) Plasma-treated
725 plastic substrate [111].

726
727

728 Zhang et al. [112] combined roll to roll and inkjet printing techniques together to develop a film-shaped
729 TEG. To specify, the authors used single-walled carbon nanotubes (SWCNT) /PEDOT:PSS
730 composites and SWCNTs/ triphenylphosphine for the *p*- and *n*-type legs, respectively. Using roll to
731 roll printing, PEDOT: PSS ink was deposited onto a polyimide substrate to serve as electrodes.
732 Notably, the PEDOT: PSS films (possessed 20 mm length each) were printed with 20 mm intervals.
733 Then, following the length and intervals of the electrodes, a PDMS film was prepared and pierced with
734 holes with 20 mm intervals. Next, the pierced PDMS film was placed on the PEDOT: PSS printed
735 substrate, while its holes were aligned with the two ends of the electrodes. After that, using inkjet
736 printing, the holes were alternately filled with the *p*- and *n*-type TE materials followed by drying them
737 on a hotplate. Finally, the holes were covered by another PEDOT: PSS printed polyimide substrate as
738 the upper electrode, see Fig. 26. The results revealed that the fabricated TEG comprised of 10
739 thermocouples obtained the maximum power density of $0.05 \mu\text{W}/\text{cm}^2$ at a temperature gradient of
740 20°C .

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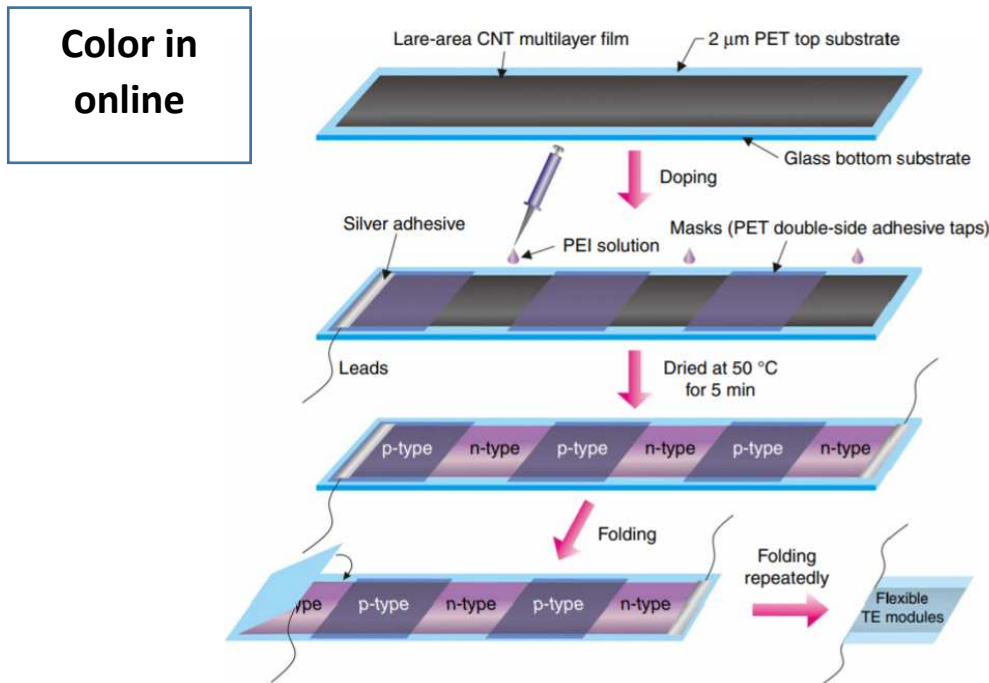
743 **Fig. 26.** Demonstration of the applied roll to roll and inject printing techniques to fabricate the film-shaped TEG [112].

744

745 Obtaining an air-stable and high performance *n*-type TE material is recognized as an important and
746 challenging topic in the TE field [113]. This is mainly because, *n*-type organic materials are mostly
747 prepared by chemical doping, resulting in their highly volatility upon air exposure. Due to the
748 correlation between the air stability of the *n*-type TE materials and their power factor, their air stability
749 is evaluated by the change in their power factor over time [114]. Accordingly, to generate a TEG
750 possessing an air-stable and a high power factor *n*-type material, Zhou et al. [115] proposed developing
751 a film-based TEG via alternately converting a pristine *p*-type film to an *n*-type one. Accordingly, the
752 authors developed a *p*-type carbon nanotube (CNT) film with the dimensions of $16 \text{ mm} \times 10 \text{ mm} \times$

753 0.15 mm onto a glass substrate. Then the film was mask shaded in a strip pattern using polyethylene
 754 terephthalate (PET) double-sided adhesive tapes, see Fig. 27. Then, the *n*-type 1wt.%
 755 polyethylenimine (PEI) solution was dropped onto the unshaded parts of the *p*-type film. After treating
 756 the doped film for 5 min at 50°C, the adhesive tapes were removed and the prepared TEG was
 757 repeatedly folded. The results showed that three pairs of this continuous TE films obtained the
 758 maximum power density of 0.06 $\mu\text{W}/\text{cm}^2$ at ΔT of 19°C. Furthermore, the variations of electrical
 759 conductivity and Seebeck coefficient were less than 5% during 3 months.

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Fig. 27. The processes of fabricating the foldable TE module [115].

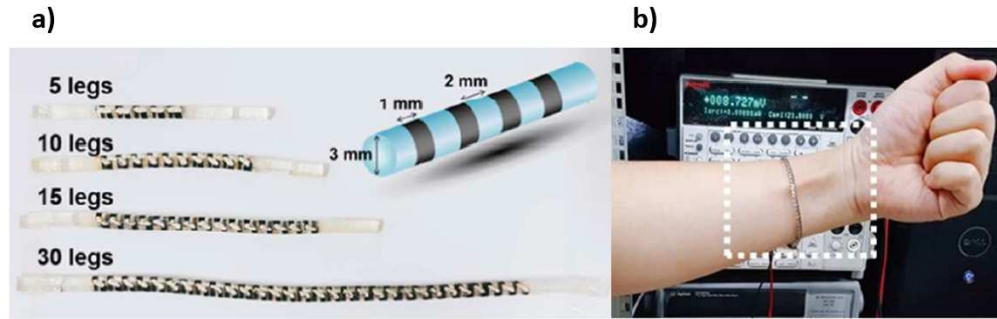
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764 Instead of a bipolar film-shaped TEG, Wang et al. [116] developed a unipolar one by drop casting *p*-
 765 type 65wt% SWNTs/polyaniline (PANI) solution onto a glass substrate. To illustrate, initially the
 766 substrate was mask shaded in a strip pattern followed by dropping the composite solution and drying
 767 it at 408°C. Finally, the four *p*-type legs each with the dimensions of 22 mm × 10 mm × 0.15 mm were
 768 connected together using silver paste and gold wires as electrodes. The results showed that the
 769 developed TEG generated the maximum output power of ~0.012 $\mu\text{W}/\text{cm}^2$ at ΔT of 30°C.

770 Stepien et al. [117] replaced the drop casting technique with dispenser printing to fabricate *p*-type film-
771 shaped legs onto a glass substrate. Regarding the dispenser printing, a nozzle dispensed continuous
772 filament of *p*-type PEDOT: PSS ink onto the substrate without any mask shading. Then, the 61 printed
773 TE legs were connected together with silver paste as electrodes. The results showed that at a
774 temperature gradient of 90°C, the maximum recorded output power of the TEG was 0.025 $\mu\text{W}/\text{cm}^2$.
775 Likewise, Liu et al. [118] electrochemically inkjet printed *n*-type poly[Kx(Ni-ethylenetetrathiolate)]
776 film onto a PET substrate. Then the 108 TE films were connected electrically together using gold
777 interconnections. The results demonstrated that the highest output power was 577.8 $\mu\text{W}/\text{cm}^2$ at 12°C
778 temperature gradient.

779 Some studies developed cross-plane TEGs via fabricating the organic film-shaped legs onto a 3D
780 supporting structure. Thus, these studies benefited both from the flexibility aspect of the organic TE
781 materials and the greater temperature difference between the skin and the ambient air. For example,
782 Park et al. [119] developed a bracelet TEG via automate printing of TE inks onto a flexible
783 polyurethane cable. In particular, the authors doped separately CNT ink with poly (acrylic acid) (PAA)
784 and poly(ethylenimine) (PEI) to prepare the *p*- and *n*-type inks, respectively. Then, the PAA doped
785 CNT ink was printed in a strip pattern onto one half of the polyurethane cable (having 3mm diameter)
786 served as the flexible supporting structure, see Fig. 28. After drying the printed *p*-type ink at 85°C
787 vacuum for 1hr, the *n*-type ink was printed onto the other half of the cable with the same pattern and
788 dried overnight under the same condition. It should be noted that the legs were printed with 1mm width
789 and with 2mm intervals. Finally, the cured *p*- and *n*-type inks were electrically connected together via
790 dispenser printing of silver pastes as electrodes. The results showed that the generated springy-shaped
791 TEG comprising 60 thermocouples obtained the maximum output power of 0.2 μW and 1.95 μW at
792 ΔT of 10°C and 30°C, respectively.

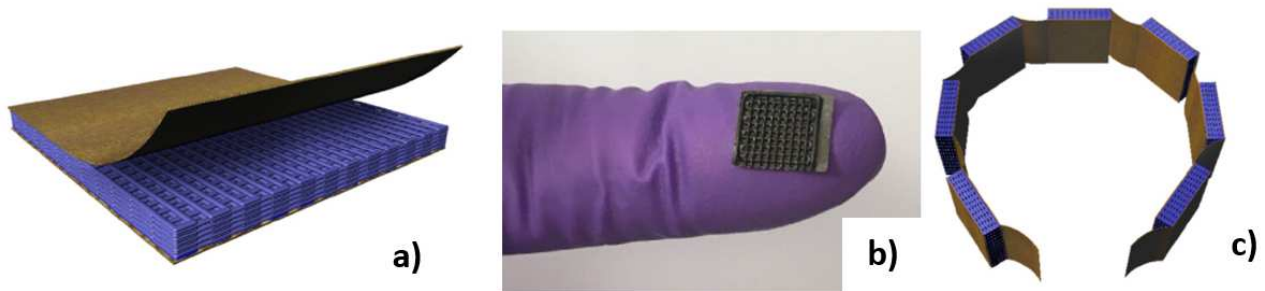
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 796 **Fig. 28.** a) Photo of the fabricated TEG with 5, 10, 15 and 30 p - n pairs; b) Demonstration of the TE bracelet wrapped
 797 around the wrist [119].

798 To develop a cross-plane TEG based on film-shaped legs, Zeng et al. [120] coated seven 3D PDMS-
 799 based grids with an organic TE material and connected them together with conducting fabrics as
 800 electrodes, see Fig. 29. To specify, the authors directly 3D printed each PDMS grid onto a Cu/Ni coated
 801 conducting fabric served as the integrating band and the bottom electrode, respectively. Then, the
 802 PDMS-based grid ($10\text{ mm} \times 10\text{ mm} \times 2\text{ mm}$) was coated with a very thin copper film via radio
 803 frequency sputtering followed by being immersed into graphene oxide dispersion as a p -type TE
 804 material. To evenly coat the grid with the TE material, the immersed substrate was subsequently coated
 805 by reduced graphene oxide (rGO) nano-sheets. After that, the top side of the developed TEG was
 806 covered with another conducting fabric as the upper electrode. The results showed that the fabricated
 807 TEG generated the output voltage of 57.33 mV/g at a temperature difference of 50°C . In addition,
 808 integrating 7 units of the developed grid-based TEG together generated the maximum power density
 809 of $4.19\text{ }\mu\text{W/g}$ at a temperature difference of 15°C .

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815
 816 **Fig. 29.** a) Schematic of the 3D printed PDMS grid; b) Photo of the fabricated grid-based TEG; c) Schematic design of
 817 integrating 7 units of the developed grid-based TEGs together [120].

818
819
820 Likewise, Liu et al. [121] developed a cross-plane TEG based on film-shaped legs, but the authors
821 utilized the electrokinetic effect rather than the Seebeck effect to harvest the thermal energy and
822 convert it into electrical energy. To specify, the authors developed a hollow cuboid bracelet as a
823 wearable TEG enclosed with an aluminium sheet from the top and bottom. The other four sides of the
824 cuboid bracelet were covered with a transparent sideboard (from the lengths) and a thermal insulator
825 (from the widths). In the hollow, the hot substrate (i.e. the bottom aluminium sheet) was covered with
826 an insulation layer to tune the heat transfer from the skin into the bracelet. As part of the TE generation
827 process, the bracelet was partially filled with deionized water from the widths.

828 To generate electricity, a porous carbon film consisted of a carbon film and interconnected carbon
829 nanoparticles was mounted on the insulation layer of the hot substrate, while its two ends were
830 immersed into the deionized water. Due to the temperature difference between the hot and cold
831 substrates, a vapour pressure gradient occurred inside the bracelet, resulting in the evaporation of the
832 absorbed water in the carbon film. After facing the cold substrate, the escaped vapour condensed and
833 flowed back to the water reservoir to repeat the cycle. Therefore, this closed cycle constantly generated
834 electricity based on water flow through the porous carbon film. The results revealed that at 13°C
835 temperature gradient, the TE bracelet generated the highest output voltage of 1V. It should be noted
836 that to guarantee a constant temperature difference, an aluminium heatsink in the form of plate fins
837 were attached to the cold substrate (i.e. top aluminium sheet) and the spaces between them were filled
838 with phase change materials.

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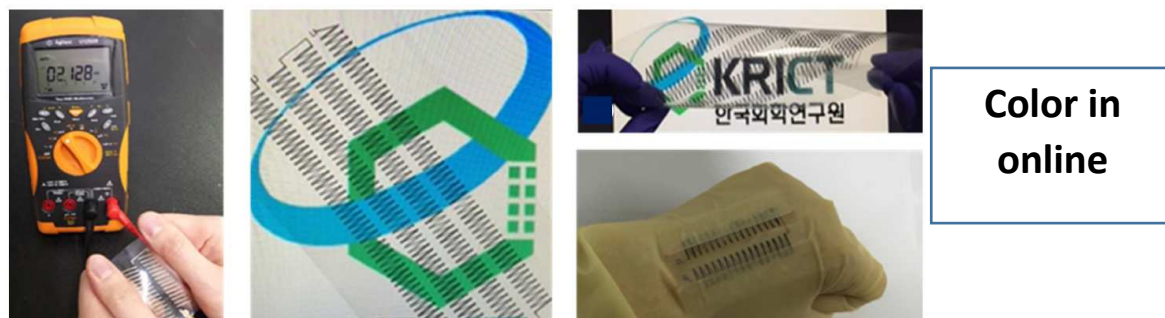
841 4.3. *Hybrid TE materials*

842

843 Inorganic TE materials benefit greatly from excellent Seebeck coefficients but suffer from high thermal
844 conductivities and brittleness. Regarding organic TE materials, they take advantage from low thermal
845 conductivities and proper film-forming performance, they but suffer from extremely low power factors
846 and stability issues [122]. Thus, it is expected to make up the shortages of these two types of TE
847 materials by combining them together. To put it another way, hybrid TE materials comprised of
848 inorganic nanostructures in a matrix of conducting polymer take advantage from both the inherently

849 low thermal conductivity of the polymer and the high power factor of the inorganic filler [123]. The
850 promising methods for preparing hybrid TE materials include physical mixing, in situ polymerization,
851 in situ synthesis, ex-situ synthesis, and layer-by-layer self-assembling [124-126]. As mentioned before,
852 power factor is a critical parameter for assessing the performance of a TEG. Thus, so far, extensive
853 research has been conducted on fabricating hybrid TE materials possessing high power factor. For
854 example, Bae et al. [127] applied H₂SO₄ chemical treatment to improve the power factor of Te-
855 PEDOT: PSS as the hybrid TE material of a unipolar film-shaped TEG. To illustrate, initially, the *p*-
856 type hybrid TE material was spin-coated onto a flexible PET (30mm × 10mm) substrate to fabricate
857 32 film-shaped TE legs, see Fig. 30. Next, the power factor of the Te-PEDOT: PSS films were
858 improved by being immersed in H₂SO₄ solution. Then, the conductive silver paste was printed onto the
859 glass substrate to electrically connect the legs together. The results showed that the TEG generated the
860 highest output power of 0.7 nW/cm² at ΔT of 10°C.

861



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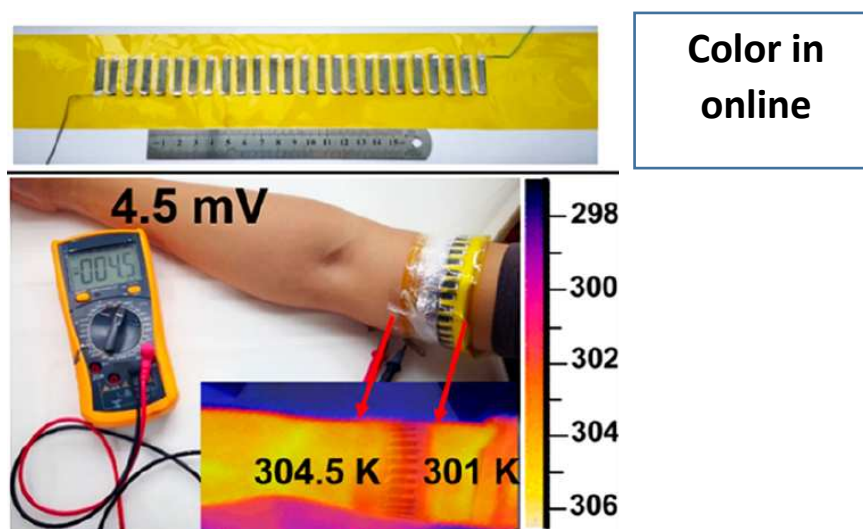
863 **Fig. 30.** Photos of the flexible and twistable TE generator comprising 240 legs arranged in four rows [127].
864

865 Likewise, Bae et al. [128] employed H₂SO₄ chemical treatment to improve the TE properties of the
866 Te-Bi₂Te₃/PEDOT:PSS composites. Using a stencil comprised of 6 rectangular openings, the
867 generated *p*-type TE ink was spray-printed onto a glass substrate. Next, the printed legs were
868 electrically connected together using silver paste. The results showed that at ΔT of 10°C, the fabricated
869 TEG generated the maximum output voltage of 1.54 mV.

870 However, in another study, Bae et al. [129] replaced the H₂SO₄ chemical treatment with adding a
871 small amount of nanocarbon material to the hybrid composite. The two selected nanocarbon materials
872 were graphene nanoparticles (GNPs) and small bundled single-walled carbon nanotubes (SSWNTs).

873 Thus, these two additives were individually added to the *p*-type hybrid Te-PEDOT: PSS composite.
874 Then, the two obtained hybrid composites were separately spray-printed onto a glass substrate to
875 fabricate 14 TE legs. Next, the legs were electrically connected together using silver paste as
876 electrodes. The results revealed that at ΔT of 20°C , the TEGs comprised of 8 wt% GNPs -Te-PEDOT:
877 PSS and 3wt% SSWNTs-Te-PEDOT: PSS composites obtained the maximum output powers of 1.7
878 nW/cm^2 and $31.5 \text{ nW}/\text{cm}^2$, respectively.

879
880 Regarding hybrid TE materials, since an inorganic TE filler usually shows higher TE properties than
881 the polymer matrix, superior TE properties could be achieved by increasing the inorganic TE
882 component. Accordingly, in situ synthesis is one of the most effective technique to adequately disperse
883 the inorganic nanoparticles in the polymer matrix. Precisely, in this technique, the inorganic
884 nanoparticles are grown inside the polymer matrix using corresponding precursors [130]. Lu et al.
885 [126] applied this technique to grow polycrystal Cu_xSe_y (PC- Cu_xSe_y) nanowires inside the PEDOT:
886 PSS matrix. Then, the prepared flexible PEDOT:PSS/ Cu_2Se nanocomposite films on nylon
887 membranes was cut into 25 legs ($25 \text{ mm} \times 5 \text{ mm}$). Next, the *p*-type legs were pasted onto a polyimide
888 substrate with $\sim 5 \text{ mm}$ intervals and electrically connected together with silver paste, see Fig. 31. The
889 results showed that the developed prototype generated the maximum voltage of 4.5 mV at a
890 temperature difference of 6.5°C .

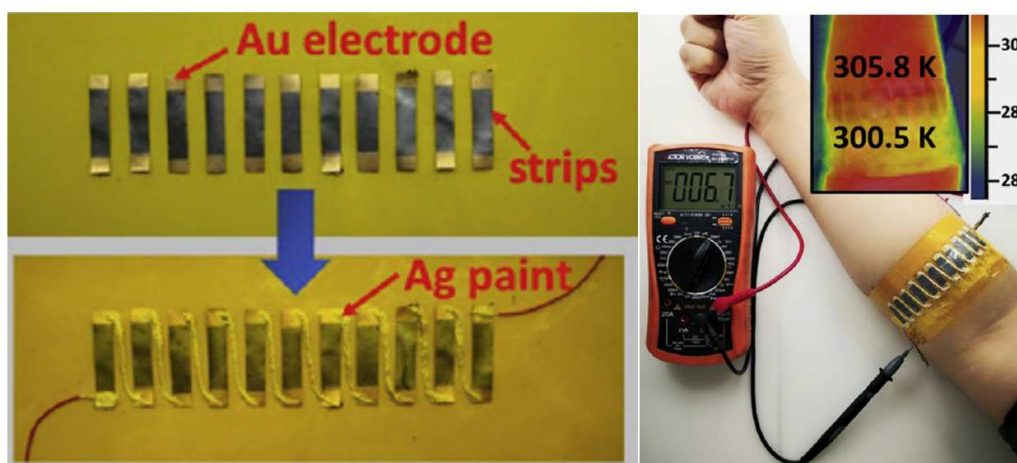


891
892 **Fig. 31.** Photos of the developed TEG and the 4.5 mV voltage created at 3.5°C temperature difference [126].
893

894 Among all the conducting polymers and inorganic TE materials at room temperature, PEDOT:PSS and
895 BiTe based alloys show the highest TE properties, respectively. However, mixing them together (i.e.

896 BiTe based alloy/ PEDOT:PSS nanocomposites) results in a lower TE properties than each one. This
897 is owing to the easy oxidation of BiTe based alloy nanostructures and its difficult dispersion in the
898 PEDOT:PSS matrix [131]. Addressing these two issues, Du et al. [131] initially exfoliated the BiTe
899 based alloy particles into nanosheets. Next, the BiTe based nanosheets/ PEDOT:PSS nanocomposites
900 was prepared on nylon membranes by vacuum assisted filtration followed by hot pressing. Then, the
901 prepared *p*-type TE film was cut into five strips (25 mm × 5 mm) followed by being adhered onto a
902 polyimide substrate with ~ 5 mm intervals. Next, the *p*-type TE strips were connected together using
903 silver paint as electrodes. The authors demonstrated that the developed TEG generated 16.9 nW at
904 47.2°C temperature gradient.

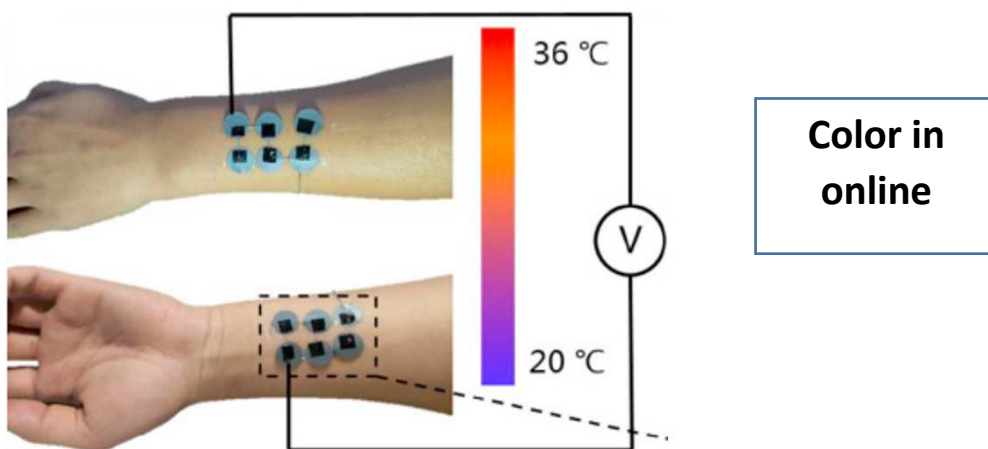
905 To homogeneously disperse the inorganic TE nanostructures in the polymer matrix, Lu et al. [132]
906 combined in situ synthetization with vacuum assisted filtration. Precisely, the authors prepared *n*-type
907 PEDOT/Ag₂Se/CuAgSe composite film by initially growing Ag₂Se/CuAgSe nanowires in PEDOT:
908 PSS. Next, the PEDOT/Ag₂Se/CuAgSe composite was prepared on porous nylon membranes by
909 vacuum assisted filtering followed by hot pressing. After that, the *n*-type film was cut into 11 legs (25
910 mm × 5 mm) and pasted onto a polyimide substrate with ~5 mm intervals. Then, the two ends of each
911 leg were masked with a layer of gold evaporation as electrodes, see Fig. 32. Next, the electrodes were
912 connected together by painting silver adhesive from one end of each strip to the opposite end of the
913 next closest strip. The authors demonstrated that the maximum power density of the fabricated TEG
914 was about 840 μW/cm² at a temperature difference of 36°C.



915
916 **Fig. 32.** Photos of the fabricated TEG and its open-circuit voltage after being placed on the arm [132].
917
918

919 However, due to the aging potential and poor biocompatibility of polymers, Zhao et al. [133] proposed
920 replacing the polymer matrix with cellulose nanofibers (CNFs). Regarding cellulose, it is a great
921 substitute for synthetic polymers owing to be abundant, renewable, and natural polymer [134].
922 Accordingly, Zhao et al. [133] added a small quantity of cellulose nanofibers into *p*-type $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$
923 and *n*-type $\text{Bi}_2\text{Se}_{0.3}\text{Te}_{2.7}$ powders, resulting in a stable and uniformly dispersed suspension. Then, a
924 vacuum filtration method was applied to prepare the self-supporting *p*-type (CNFs/ $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$) and
925 *n*-type (CNFs/ $\text{Bi}_2\text{Se}_{0.3}\text{Te}_{2.7}$) films. After that, 12 circles (each with the diameter of 5mm) were cut out
926 of the two films in total followed by arranging them in two groups of six on both sides of the arm, see
927 Fig. 33. Then, the TE films were connected together by a silver wire as an electrode. Notably, a silver
928 paste was deposited at the junctions of the wire and TE films to reduce their contact resistance.
929 Ultimately, a carbon conductive tape firmly attached the wire to the films. The results showed that
930 when the TEG was tested at ΔT of 35°C , it generated the highest output voltage of 6.3mV.

931
932



933
934
935 **Fig. 33.** Photo of the fabricated TEG and its mounting position on the arm [133].
936
937

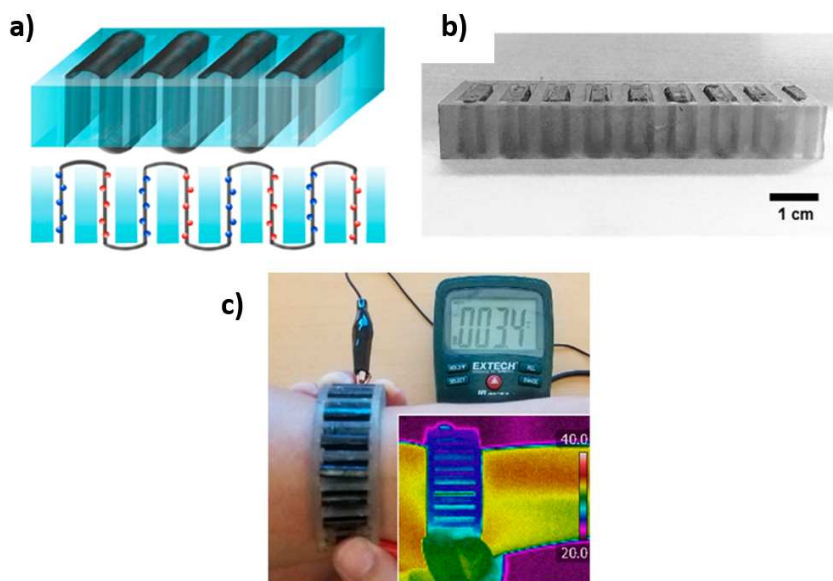
938 Another approach to improve the electrical conductivity and flexibility of hybrid films is using
939 conductive skeletons. To clarify, conductive skeleton performs as a bridge to connect large nanoplates
940 together. Accordingly, Wu et al. [135] used the reduced graphene oxide (RGO) and SWCNTs networks
941 as the conductive skeletons of Bi_2Te_3 and Sb_2Te_3 nanoplates, respectively. Then, the as-assembled
942 flexible *n*-type (RGO/ Bi_2Te_3) and *p*-type (SWCNTs/ Sb_2Te_3) films were cut into ten uniform strips (30
943 mm \times 3 mm). Next, the TE strips were alternately fabricated onto a one-sided adhesive polyimide tape
944 as the underlying substrate followed by being electrically connected together with a highly conductive

945 silver paste. To isolate the skin from the potential toxicity of the carbon nanomaterials, the air exposed
946 side of the TEG was covered with a soft polyamide film. The results proved that the fabricated TEG
947 comprising 10 thermocouples obtained $\sim 0.7 \mu\text{W}/\text{cm}^2$ at a temperature gradient of 70°C .

948 Instead of fabricating the film-shaped hybrid legs onto a flat underlying surface, some researches
949 attempted to fabricate them onto a 3D substrate to take advantage from a greater temperature difference
950 along/across the legs. For example, Xu et al. [136] developed a helical TE bracelet via brush printing
951 TE pastes onto a helical polymer substrate. Precisely, the authors obtained a viscous polymer solution
952 by dissolving Polyurethane (PU) in *N,N*-dimethylformamide. Then, a screw was fully coated with the
953 as-prepared polymer solution and dried naturally to develop a helical polymer substrate. To prepare
954 the TE pastes, Bi_2Te_3 (*p*-type) and Bi_2Se_3 (*n*-type) powders were separately mixed with a homemade
955 polyvinylidene fluoride (PVDF) binder. Next, the pastes were split-brushed onto the helical polymer
956 substrate such that they filled the pitches of the screw. After drying the TE films at 80°C for 15 min,
957 they were electrically connected together via a conductive silver paste. When the silver paste was cured
958 in an 80°C oven for about 10 min, the fabricated TEG was peeled off the screw. The results showed
959 that wearing the helical TEG on the arm generated the highest output voltage of 8.9mV at an ambient
960 temperature of $\sim 16^\circ\text{C}$.

961 Rather than printing the film-shaped legs onto a 3D substrate, Choi et al. [137] embedded a hybrid film
962 sheet within a 3D substrate. To illustrate, a highly aligned CNT sheet ($200 \text{ mm} \times 15 \text{ mm}$) was doped
963 alternately with FeCl_3 (*p*-type) and Polyethylenimine (*n*-type) solutions. Notably, the *p*- and *n*-type
964 solutions were doped with 2 mm intervals and the un-doped areas performed as electrodes to connect
965 the legs electrically together. Finally, the doped CNT sheet comprising 9 continuous thermocouples
966 was embedded in a pre-prepared PDMS block, see Fig. 34. Accordingly, the *p*- and *n*- parts of the CNT
967 sheet were embedded in the PDMS substrate, leaving the un-doped regions (i.e. electrodes) exposed to
968 the ambient air. The results revealed that when the fabricated prototype was worn on the wrist, it
969 achieved 3.4mV at ΔT of $\sim 7^\circ\text{C}$.

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973 **Fig. 34.** a) Schematic structure of the developed prototype; b) Photo of the prototype; c) Demonstration of the prototype
 974 fabricated on the arm [137].
 975

976 Instead of preparing hybrid films, some researches combined the electrically opposite inorganic and
 977 organic legs together. For example, Na et al. [138] electrodeposited *p*-type PEDOT and *n*-type Bi₂Te₃
 978 films onto a fluorine-doped tin oxide (FTO) glass substrate and a stainless steel substrate (SSS),
 979 respectively. Then the electrodeposited films were detached from the FTO and SSS substrates using
 980 an adhesive tape. Finally, the 10 adhered TE films (each with the dimensions of 5 mm × 15 mm) to the
 981 adhesive tape were connected together with a layer of gold evaporation as electrodes. The results
 982 proved that at ΔT of 12°C, the highest power density generated by the fabricated TEG was 3.45
 983 nW/cm².

984

985 Likewise, Du et al. [139] integrated *p*-type organic legs with *n*-type inorganic ones, but the main
 986 difference was that only the *p*-type legs were film-shaped and the *n*-type ones comprised of wire. To
 987 specify, the authors fabricated an air-permeable and wearable TEG via combining PEDOT: PSS coated
 988 films (*p*-type) with Constantan wires (*n*-type). Accordingly, the *p*-type legs were produced by coating
 989 a commercial cotton fabric with PEDOT: PSS followed by cutting it into five strips (each strip was 35
 990 mm long and 5 mm wide). Using silver paint as a *p*-type adhesive, the *p*-type strips were adhered with
 991 6 mm intervals to another cotton fabric. Finally, the strips were connected together with a Constantan
 992 wires served both as the electrodes and the *n*-type legs. The results demonstrated that the generated

993 TEG obtained 10.63 nW/cm^2 at ΔT of 74.3°C . [Table 3](#) illustrates the summary of the reviewed
994 literatures on wearable TEGs consisting film-shaped legs.

995

996

997

998 **Table 3.** Summary of the recently developed wearable TEGs comprising film-shaped thermocouples.

<i>P</i> -type	<i>N</i> -type	Couple Number	Electrode	Substrate	ΔT (°C)	Output voltage or power	Ref
<i>Inorganic</i>							
n/a	Bi ₂ Te _{2.8} Se _{0.2}	5	Copper foils	Polyimide substrate	20	500 $\mu\text{W}/\text{cm}^2$	[98]
Au-MoS ₂ film	n/a	5	Metal yarn	Fabric	5	2.5mV	[100]
Sb ₂ Te ₃	Bi _{1.8} Te _{3.2}	8	SbTe	Polyimide substrate	20	0.016 $\mu\text{W}/\text{cm}^2$	[101]
Bi _{0.5} Sb _{1.5} Te ₃	Bi ₂ Te ₃	5	EGaIn	Polyimide substrate	7	14.1 nW	[103]
SnTe	PbTe	32	Aluminium films	Polyimide substrate	120	8.5 mW/cm^2	[105]
Bi _{0.5} Sb _{1.5} Te ₃	Bi ₂ Te _{2.8} Se _{0.3}	56	Au-Ge	Polyimide substrate	93	18.62 $\mu\text{W}/\text{cm}^2$	[106]
Sb ₂ Te ₃	Bi ₂ Te _{2.7} Se _{0.3}	10	n/a	Polyimide substrate	20	0.095 $\mu\text{W}/\text{cm}^2$	[107]
<i>Organic</i>							
PEDOT: PSS: DMSO	Poly[Na(NiETT)]	16	Silicone-based silver ink	Polyester knitted fabric	3	3mV	[110]
PEDOT: PSS	Nitrogen-doped graphene	18	PEDOT: PSS	Plastic substrate	10	0.024 $\mu\text{W}/\text{cm}^2$	[111]
SWCNT/PEDOT:PSS	SWCNTs/triphenylphosphine	10	PEDOT: PSS	Polyimide substrate+ PDMS film	20	0.05 $\mu\text{W}/\text{cm}^2$	[112]
CNT film	polyethylene terephthalate (PET)	3	n/a	Glass substrate	19	0.06 $\mu\text{W}/\text{cm}^2$	[115]
SWNTs/ PANI	n/a	4	Silver paste	Glass substrate	30	0.012 $\mu\text{W}/\text{cm}^2$	[116]
PEDOT: PSS	n/a	61	Silver paste	Glass substrate	90	0.025 $\mu\text{W}/\text{cm}^2$	[117]
n/a	poly[Kx(Ni-ethylenetetrathiolate)]	108	Gold	PET substrate	12	577.8 $\mu\text{W}/\text{cm}^2$	[118]
poly (acrylic acid) doped CNT	poly(ethylenimine) doped CNT	60	Silver paste	Flexible cable	30	1.95 μW	[119]
Graphene oxide	n/a	7	Cu/Ni coated conducting fabric	PDMS grid	15	4.19 $\mu\text{W}/\text{g}$	[120]
<i>Hybrid</i>							
PEDOT: PSS coated PC-Cu _x Se _y film	n/a	25	Silver paste	Polyimide substrate	6.5	4.5 mV	[126]
Te-PEDOT:PSS	n/a	32	Silver paste	PET substrate	10	7e-4 $\mu\text{W}/\text{cm}^2$	[127]
Te-Bi ₂ Te ₃ /PEDOT:PSS	n/a	6	Silver paste	Glass substrate	10	1.54 mV	[128]
SSWNTs-Te-PEDOT: PSS	n/a	14	Silver paste	Glass substrate	20	0.031 $\mu\text{W}/\text{cm}^2$	[129]
BiTe /PEDOT:PSS	n/a	5	Silver paint	Polyimide substrate	47.2	16.9 nW	[131]

n/a	PEDOT/Ag ₂ Se/CuAgSe	11	Gold	Polyimide substrate	36	840 $\mu\text{W}/\text{cm}^2$	[132]
CNF/Bi _{10.5} Sb _{1.5} Te ₃	CNF/Bi ₂ Se _{0.3} Te _{2.7}	6	Silver wire	n/a	35	6.3mV	[133]
SWCNTs/Sb ₂ Te ₃ film	RGO/Bi ₂ Te ₃ film	10	Silver paste	Polyimide substrate	70	0.7 $\mu\text{W}/\text{cm}^2$	[135]
Bi ₂ Te ₃ /PVDF	Bi ₂ Se ₃ /PVDF	-	Silver paste	Helical polymer substrate	16	8.9mV	[136]
FeCl ₃ doped CNT	Polyethylenimine doped CNT	9	CNT	PDMS	7	3.4mV	[137]
PEDOT	Bi ₂ Te ₃	5	Gold	Adhesive film substrate	12	0.003 $\mu\text{W}/\text{cm}^2$	[138]
PEDOT: PSS coated cotton fabric	Constantan wires	5	Constantan wires	TE-strips attached to a cotton fabric using silver paint	74.3	10.63 nW/cm ²	[139]

999

1000 5. Yarn-shaped thermocouples

1001

1002 5.1. Inorganic TE materials

1003

1004 As reviewed in section 4, film-shaped TE legs generate considerably lower output power than the
1005 ingot-shaped ones. This is mainly because the former harvests the thermal energy in the in-plane (i.e.
1006 length/width) direction, while the latter utilizes the perpendicular temperature gradient formed between
1007 the human body and the environment. However, the main privilege of the film-shaped legs over the
1008 ingot-shaped ones is their 2D architecture, making them compatible with the curved surfaces of the
1009 body. Accordingly, yarn-shaped legs have been developed to take advantage from both the temperature
1010 gradient in the thickness direction and flexibility. Regarding the perpendicular temperature difference,
1011 a 3D flexible substrate is required to hold thin TE yarns/threads/fibres in 3D space (i.e. upright in the
1012 practical direction of the heat flow). For example, Lee et al. [140] attempted to provide an appropriate
1013 thickness for a TE yarn by specifying three different weaving patterns for it, including zigzag weave,
1014 garter-stitch weave, and plain weave. Based on these three specified weaving patterns, the authors
1015 developed three different TE yarns, including *p*- type yarn, *n*-type yarn, and the yarn containing
1016 alternate *p*- and *n*-type segments (i.e. integral *p-n* yarn). To specify, the individual *p*- and *n*-type yarns
1017 were developed by separately coating electrospun polymer nanofiber cores with *n*- (Bi₂Te₃) and *p*-
1018 (Sb₂Te₃) type semiconductor sheaths, using radio-frequency-magnetron sputtering technique. Then the

1019 coated cores were twisted into separate *n*- and *p*-type yarns. For the integral *p-n* yarn, the authors
1020 alternately sputtered strips of Sb_2Te_3 and Bi_2Te_3 on both sides of the electrospun polymer nanofiber
1021 cores. Furthermore, narrow gold strips were sputtered at the intervals of the *p*- and *n*-type segments to
1022 electrically connect them together. After preparing the TE yarns, the separate *p*- and *n*-type yarns were
1023 used for fabricating the zigzag and garter-stitch weaves, whereas the integral *p-n* yarn was used for
1024 developing the plain weave. The results demonstrated that at ΔT of 20°C , the approximate output
1025 power of the zigzag, garter-stitch, and plain weaving patterns were 0.02 W/m^2 , 0.01 W/m^2 , and 0.08
1026 W/m^2 , respectively.

1027
1028 Apart from providing an appropriate thickness, another approach to improve the efficiency of yarn-
1029 based TEGs is applying highly efficient TE materials. To illustrate, Zhang et al. [141] prepared TE
1030 fibres with single-crystal SnSe rods owing to their superior mechanical stability, low optical loss, and
1031 excellent electrical conductivity. Precisely, *p*-type multicore SnSe fibres were prepared by cladding
1032 several single-crystal SnSe rods with borosilicate glass and borosilicate glass rods. Then, 6 multicore
1033 SnSe fibres were weaved into a shirt followed by holding them in place by depositing silver films at
1034 their two ends. Finally, the TE fibres were serially connected together with copper wires as electrodes.
1035 The results illustrated that the TE shirt delivered 30 mV at ΔT of 10°C .

1036

1037 5.2. *Organic TE materials*

1038

1039 Although inorganic TE materials can be used for fabricating TE yarns, their intrinsic rigidity limits the
1040 flexibility of the yarns. Therefore, it is prevalent to apply organic or hybrid TE materials in the
1041 fabrication of TE yarns. Furthermore, as mentioned before, using a 2D flat substrate (in-plane design)
1042 restricts the output voltage/power of wearable TEGs, because it poorly fit to the perpendicular
1043 temperature gradient formed between the human body and the environment. Thus, it is desirable to
1044 consider a solution to change the 2D flat substrate into a 3D architecture. Accordingly, using either a
1045 3D spacer fabric or a thick textile is a promising candidate for holding the TE yarns upright in the
1046 direction of the perpendicular temperature gradient. Accordingly, Wu & Hu [142] utilized a 3D spacer
1047 fabric as the underlying substrate to embroider the TE yarns into it. Regarding the spacer fabric, it
1048 consisted of two separate outer fabric layers joined together but kept apart by spacer yarns. To specify,

1049 the authors introduced a fibre-shaped TEG by coating commercial polyester yarns with *p*- and *n*-type
1050 organic composites. In particular, NWPU/PEDOT: PSS/multi-walled carbon nanotube (MWCNT)
1051 and NWPU/nitrogen doped MWCNT were used as the *p*- and *n*-type TE composites, respectively.
1052 Then, the prepared TE yarns were alternately embroidered into the lock-nit surface of the 3D spacer
1053 fabric. Next, the legs were connected together from the top and the bottom with silver paint as
1054 electrode. The results exhibited that the resultant TE fabric comprising 10 thermocouples produced the
1055 maximum output power of 0.21 nW/cm² at ΔT of 66°C.

1056
1057 Instead of a 3D spacer fabric, Lund et al. [143] used a 3D textile to stitch the *p*- (PEDOT:PSS coated
1058 silk thread) and *n*-type (silver-plated polyamide thread) threads through it. To prepare the *p*-type
1059 thread, 100 m of a silk sewing thread was passed through a dye bath that was filled with aqueous
1060 PEDOT: PSS dispersion and ethylene glycol. Then the coated thread was dried and treated with
1061 dimethyl sulfoxide for 1 hr and 20 min. Finally, the coated thread was dried again at 100°C with a heat
1062 gun. Next, the TE legs were formed by stitching the TE threads through 9 layers of a felted wool fabric
1063 followed by being electrically connected together with silver paste, as Fig. 35. To cure the paste, the
1064 TE textile was placed in a 100°C oven for 10 min. The results showed that the developed TE textile
1065 comprising 8 thermocouples obtained the maximum output power of 0.04 μ W/cm² at ΔT of 65°C.

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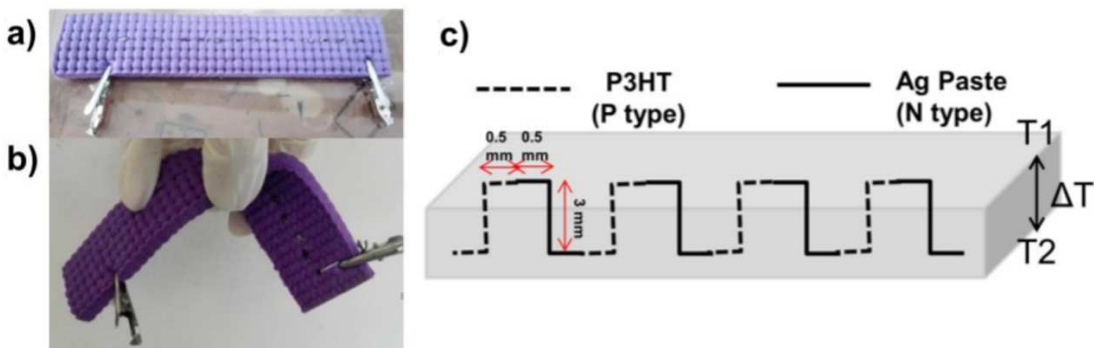
1068
1069 **Fig. 35.** Photos of the embroidered thread-based thermopile [143].

1070
1071 Qu et al. [144] replaced the separate *p*- and *n*-type threads with an integral *p-n* thread to sew it onto a
1072 3D fabric-based substrate. To illustrate, the authors developed the *p*-type thread by immersing a cotton

1073 thread in the poly(3-hexylthiophene) (P3HT) solution as the *p*-type material. Then, the P3HT coated
1074 thread was wrapped around a supporting cell block. To obtain the *n*-type thread, on half of the P3HT
1075 coated thread wrapped around the block was brushed with an Ag paste as an *n*-type material. After
1076 that, the serial P3HT /Ag coated cotton thread was released from the supporting cell and sewn onto a
1077 flexible 3D fabric, see Fig. 36. The results demonstrated that the maximum output power of the device
1078 with 13 thermocouples was 1.15 μW at a temperature gradient of 50°C.

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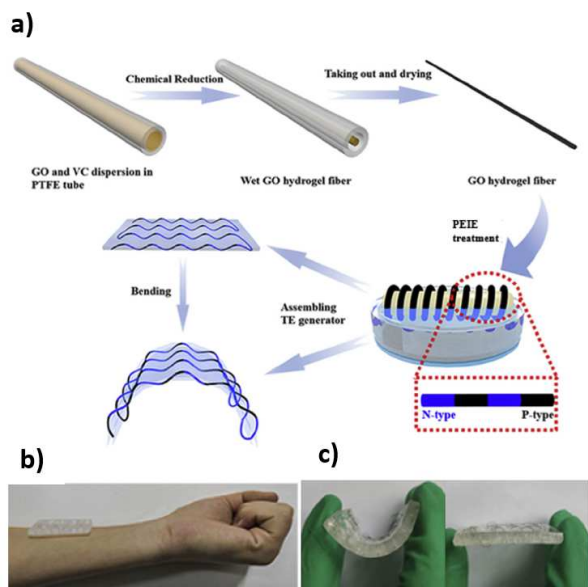
1082
1083 **Fig. 36.** a) Photo of the fabricated TEG; b) Flexibility of the fabric; c) Schematic of the TEG's structure [144].
1084

1085 Ito et al. [145] not only applied the same technique to fabricate an integral *p-n* type thread, but also
1086 used electrodes to electrically connect the *p*- and *n*-type segments together. To illustrate, a SWCNT
1087 thread was initially injected into a methanol bath and left there for 24 hrs to be converted into a *p*-type
1088 thread. Then, the wet thread was pulled out and dried, resulting in a CNT/PEG composite thread. To
1089 generate the *n*-type segments, the as prepared *p*-type CNT/PEG composite thread was wound around
1090 a long and narrow plastic plate. Then, an *n*-type doping agent called 1-butyl-3-methylimidazolium
1091 hexafluorophosphate ([BMIM]PF6) was doped onto one side of the plastic plate, converting the *p*-type
1092 thread into the *n*-type. Next, the *p*- and *n*-type segments were electrically connected together by
1093 painting conductive silver paste at their junctions. Finally, the stripe patterned thread was sewn onto a
1094 thick felt fabric having 3 mm thick and 400mm² base area. The authors revealed that at 22°C ambient
1095 temperature, the TEG obtained the maximum output power of 0.125 nW/cm² when heated by the
1096 human body (~37°C).

1097 Zheng et al. [64] applied a weft-knitted spacer fabric as the 3D substrate to directly knit the integral *p*-
1098 *n* type yarn into it. To prepare the *p-n* segmented yarn, a pristine CNT yarn was rolled onto a thin PET-
1099 based cylindrical structure. Using a silver paste, two parallel lines were drawn onto the rolled CNT
1100 yarn to divide it into three equal segments. Subsequently, the two end segments were immersed into
1101 aqueous PEDOT: PSS and PEI/ethanol solutions to form the *p*- and *n*-type legs, respectively, leaving
1102 the middle segment pristine. Finally, 966 pairs of the alternately PEDOT: PSS- CNT- PEI immersed
1103 yarn was woven into the spacer fabric. The results showed that the developed TE textile generated the
1104 maximum output power of 0.14 $\mu\text{W}/\text{cm}^2$ when it was directly put on the arm (ΔT of $\sim 7^\circ\text{C}$).

1105
1106 To provide the required thickness for the yarn-shaped legs, Lin et al. [146] replaced the 3D fabric-
1107 based substrate with a PDMS block. To illustrate, initially *p*-type graphene fibre was developed by
1108 sealing a homogeneously mixed graphene oxide and ascorbic acid solution in a polytetrafluoroethylene
1109 tube. Then, the obtained *p*-type graphene fibre was taken out of the tube followed by being chemically
1110 treated with $\text{N}_2\text{H}_4 \cdot \text{H}_2\text{O}$. Next, to obtain an integral *p-n* segmented fibre, the as-prepared *p*-type
1111 graphene fibre was rolled onto a plastic rod followed by immersing one half of the rod into the electron
1112 donating polyethyleneimine ethoxylated solution, see Fig. 37. Finally, the integral *p-n* graphene fibre
1113 comprising 20 thermocouples was woven into a PDMS block to obtain a flexible fibre-based TE
1114 device. The results recorded that the highest delivered output power was $\sim 1.3 \text{ pW}$ at ΔT of $10 \pm 0.5^\circ\text{C}$.

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1121
 1122 **Fig. 37.** a) Schematic of the fabrication processes of the integral *p-n* fibre; b) Photo of the fabricated TEG; c) Bendability
 1123 of the developed TEG [146].

1124
 1125 However, the main disadvantages of using a 3D flexible substrate for a yarn-based TEG is that they
 1126 may cause parasitic loss of temperature difference or deteriorate the flexibility. Thus, rather than using
 1127 an external object as the 3D substrate, Sun et al. [147] hold the TE yarns in the vertical direction by
 1128 specifying two different knitting patterns. To illustrate, the authors developed integral *p-n* carbon
 1129 nanotube fibres (CNTFs) by alternately coating it with active *p*- and *n*-type materials. Accordingly,
 1130 each carbon nanotube fibre was initially generated by twisting four CNT films together. Then, each
 1131 CNTF was divided into three alternate segments, including *p*-hybridized, electrode (i.e. the un-doped
 1132 CNTF), and *n*-doped segments. To fabricate the *p*-hybridized segments, CNTFs were dipped into a
 1133 commercial poly(3,4-ethylenedioxythiophene): poly(styrenesulfonate) (PEDOT: PSS) solution. To
 1134 electro spray the *n*-type oleamine solution onto the CNTFs, both the *p*-hybridized and the electrode
 1135 segments were mask shaded with a polypropylene mask. After that, the coated CNTFs were wrapped
 1136 with acrylic fibres to avoid any short circuit, see Fig. 38. Finally, the authors interlocked 15
 1137 thermocouples together in two different patterns, including warp knitting and weft knitting. The results
 1138 showed that at ΔT of 44°C, the highest output power was achieved by the warp knitting with 7 $\mu W/cm^2$.
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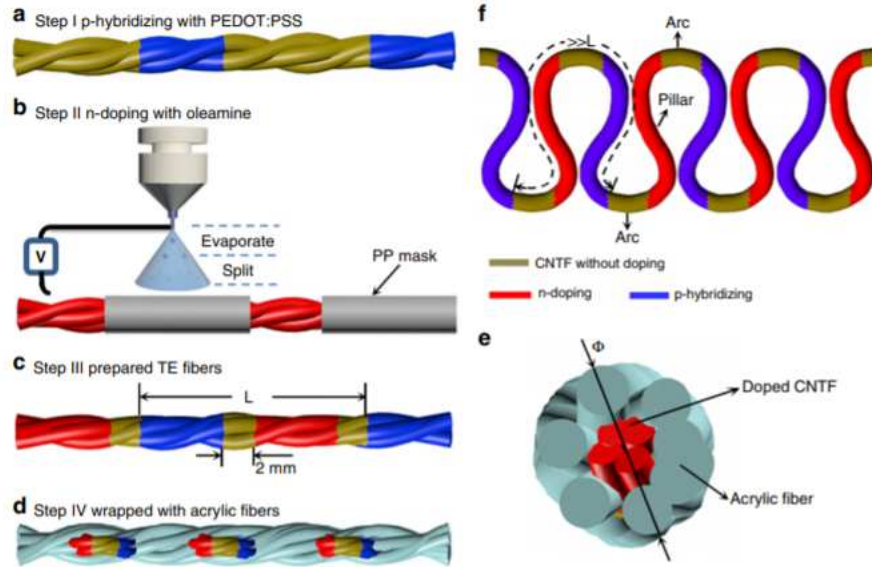


Fig. 38. Schematic of the fabrication procedure of the TE fibre [147].

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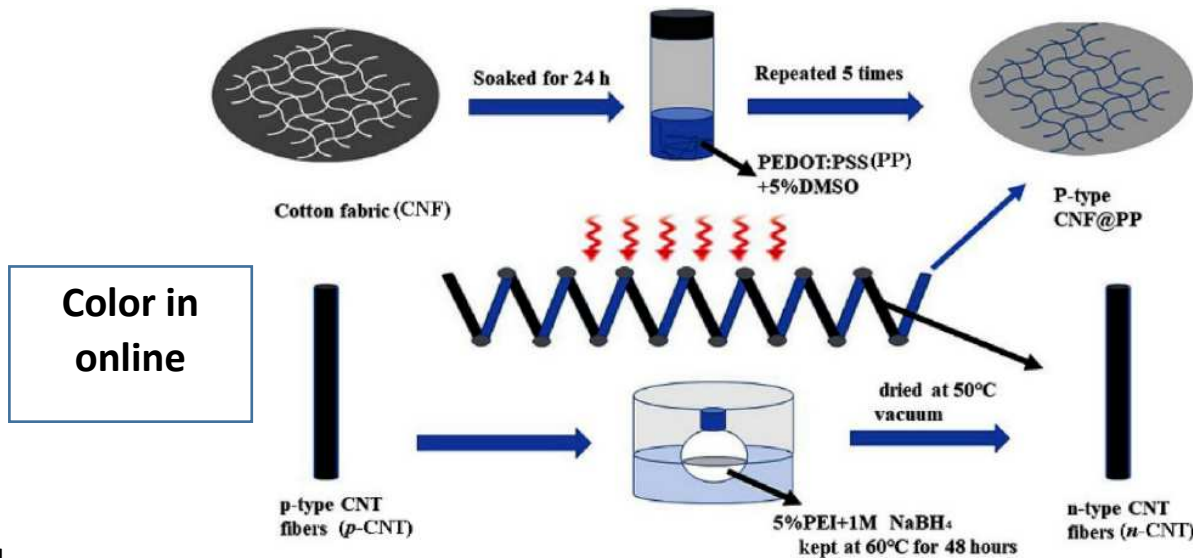
1143 Allison et al. [148] developed the 3D flexible substrate via folding a 2D flat substrate in half along its
1144 x-axis. It is noteworthy that the authors combined *p*-type film-shaped and *n*-type yarn-shaped legs
1145 together to develop their fabric-based TEG. To specify, initially *p*-doped poly(3,4-
1146 ethylenedioxythiophene) (PEDOT-Cl) was persistently vapour printed onto a commercial cotton
1147 fabric. Then, two rectangles each with the dimensions of 45 mm × 5 mm were cut out of a polyimide
1148 stencil to mask shade the vapour printed cotton fabric. Next, *p*-type PEDOT-Cl was screen printed
1149 onto the fabric through the two openings of the stencil. After that, the *n*-type carbon fibres were sewn
1150 onto the PEDOT-Cl coated fabric such that the two opposite ends of the PEDOT-Cl legs were bonded
1151 together. Using cotton thread, squares of silver coated nylon were sewn onto the junctions of the *p*-
1152 type PEDOT-Cl films and *n*-type carbon fibres to connect them electrically together. To increase the
1153 temperature gradient in the length direction, the cotton substrate was folded in half along the x-axis
1154 and then sewn onto a knitted band such that one half was exposed to the ambient air and the other half
1155 was buried under the knitted band. The authors demonstrated that the highest output power generated
1156 by the as-developed fabric-based TEG was 0.4 nW/cm² at ΔT of 30°C.

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1161 As mentioned before, apart from holding the TE yarns in the vertical direction, another approach for
1162 developing a highly efficient fabric-based TEG is applying highly efficient TE materials. In general,
1163 *n*-type organic materials (e.g. conducting polymers and CNTs) show poorer stability (i.e. air and
1164 electrical stabilities) and relatively lower TE performance than the *p*-type ones, which greatly hinders
1165 the development of the organic-based TE devices [149]. For example, carbon nanotube (CNT)
1166 represent a unique 1D carbon allotrope that its structural, electrical, and thermal properties lead to an
1167 efficient thermoelectric energy conversion [150]. In spite of the competitive performance of *p*-type
1168 CNTs in carbon-based TE materials, the work on *n*-type CNTs is very lacking due to their less stability
1169 than *p*-type CNTs. In particular, *n*-type CNTs can be obtained by the introduction of electron donating
1170 dopants. However, not only they are susceptible to be de-doped by oxygen in ambient conditions, such
1171 that their Seebeck coefficient becomes positive over time, but also they suffer from poor air stability
1172 [151-152].

1173
1174 To provide a stable *n*-type CNTs yarn, Lan et al. [153] proposed a chemical treatment to convert a *p*-
1175 type CNT yarn into an *n*-type. Accordingly, the authors used common cotton and CNT fibres to
1176 develop *p*- and *n*-type fibres, respectively. To prepare the *p*-type fibres, the cotton fibres were treated
1177 with absolute ethanol, deionized water, and DMSO followed by being soaked five times in PEDOT:
1178 PSS for 24 hrs. To obtain the *n*-type fibres, the *p*-type CNT fibres were treated with PEI, resulting in
1179 a high electrical conductivity and Seebeck coefficient with excellent environmental stability. As Fig.
1180 39 shows, 8 pairs of the as-prepared *p*- and *n*-type fibres each with 15 mm length were electrically
1181 connected together using silver paste. The results revealed that the developed TEG generated 10 μ W
1182 at ΔT of 10°C.

1183



1184
 1185 **Fig. 39.** Schematic of the fabrication processes of the CNF@PP (*p*-type) and the PEI treated CNT (*n*-type) fibres [153].
 1186

1187 Ryan et al. [154] demonstrated air-stable organic *n*-type yarns by coating commercial poly(ethylene
 1188 terephthalate) (PET) sewing threads with the nanocomposites of multiwalled carbon nanotubes
 1189 (MWNTs) and poly(N-vinylpyrrolidone)(PVP). The authors selected MWNT:PVP nanocomposites
 1190 owing to its readily air stable *n*-type behaviour. In particular, the authors developed a TE textile via
 1191 generating *n*- and *p*-type yarns separately and sewing them onto a wool fabric. To produce the *n*-type
 1192 yarns, commercial PET sewing threads were sequentially coated with poly(N-vinylpyrrolidone),
 1193 MWNTs, and polystyrene-*b*-polyisoprene-*b*-polystyrene block copolymer served as the adhesion
 1194 layer, the conducting layer, and the protection layer, respectively. To prepare the *p*-type yarns,
 1195 commercial silk yarns were dyed by being submerged in poly(3,4-ethylenedioxythiophene):
 1196 poly(styrenesulfonate) (PEDOT: PSS) solution. Then, as Fig. 40 depicts, 38 pairs of the TE yarns (with
 1197 40 mm length) were alternately sewn onto a fabric followed by being electrically connected together
 1198 with a conductive carbon-based paste. The results depicted that when the TE textile was heated and
 1199 cooled from the opposite lengths, it obtained the maximum output power of 7.1 nW at a temperature
 1200 gradient of 80°C.

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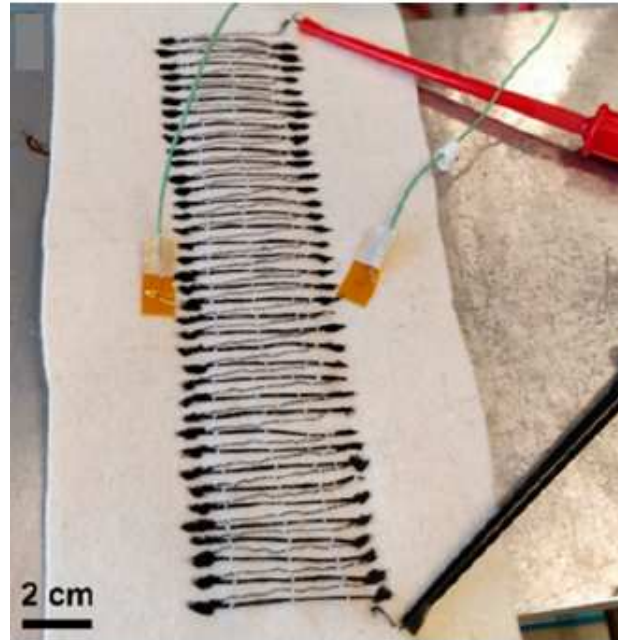


Fig. 40. All organic in-plane embroidered textile TE device with 38 n - p elements [154].

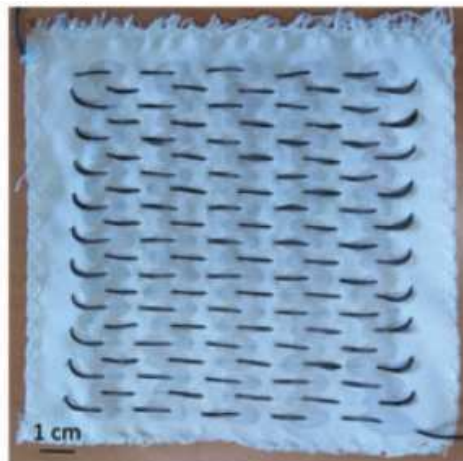
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1206 Hardianto et al. [155] replaced CNT with carbon fibre owing to its fantastic strength and lightweight.
1207 Accordingly, the authors stitched an n -type pristine carbon fibre tow onto a 120 mm \times 120 mm
1208 polyester fabric in a way that the carbon fibre floated on both sides of the polyester fabric. Notably,
1209 the length of each float was around 10 mm. As a prerequisite, roughly half of every floats on both sides
1210 of the fabric was mask shaded by acrylic dispersion to electroplate the other half with nickel particles
1211 as the p -type material, see Fig. 41. The results illustrated that the prototype with 105 carbon-nickel
1212 thermojunctions generated the highest output power of 0.041 nW/cm² at ΔT of 2°C.

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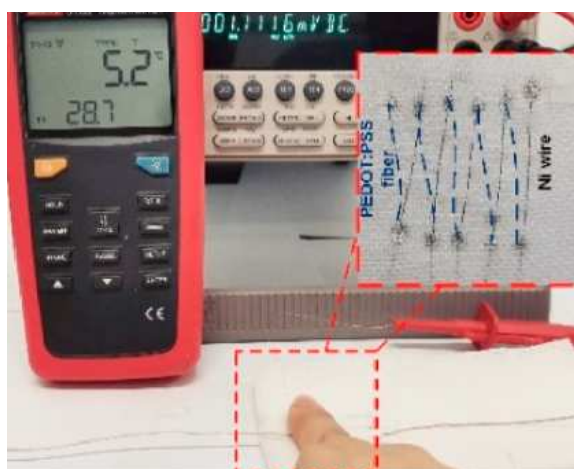
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Fig. 41. The fabricated textile-based TEG [155].

1218 Conducting polymers are another organic TE materials that have gained extensive attention for
1219 fabricating textile-based TEGs. For example, the electrically conductive polymer PEDOT:PSS has
1220 gained particular attention due to its highly conductive properties, solution processability, and
1221 remarkable easy doping [156]. In fact, the chain packing order of conducting polymers have strong
1222 impact on their TE properties [157]. For example, the power factor of a one-dimensional PEDOT:PSS
1223 fibre is 15 times greater than that of a two-dimensional PEDOT:PSS film processed at the same
1224 condition. Thus, Wen et al. [158] attempted to produce one-dimensional PEDOT:PSS fibres through a
1225 continuous wet-spinning process followed by a one-step treatment with sulfuric acid (H₂SO₄).
1226 Regarding wet spinning, polymer powder is dissolved in a suitable solvent followed by the extrusion
1227 of the polymer solution through spinning into a solvent-non solvent mixture (coagulant). After
1228 preparing one dimensional PEDOT: PSS fibres, they were sewn in diagonal pattern onto a flexible
1229 Kapton tape, resulting in 5 *p*-type legs, as Fig. 42 depicts. Then, *n*-type nickel wires (with 30 mm
1230 length) were sewn from one end of each *p*-type leg to the opposite end of the closet leg, resulting in 5
1231 thermocouples. The results showed that when one end of the fibrous TEG was touched by fingertip
1232 and the other end was exposed to the air, it delivered an open-circuit voltage of 0.72 mV at ΔT of
1233 3.6°C.

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Fig. 42. The in-plane TE power generation with fingertip, while the fibrous TEG was sewn onto a piece of cloth [158].

1238 Likewise, Kim et al. [159] combined the wet-spinning and post-treatment methods together to prepare
1239 single-walled CNT/PEDOT:PSS composite fibres. To clarify, aqueous CNTs / PEDOT: PSS paste was
1240 directly spun into methanol as the coagulation solvent. Notably, the CNTs and PEDOT: PSS content
1241 of the paste acted as the *n*- and *p*-type materials, respectively. Thus, the spun paste was extruded into
1242 the solvent and turned into the CNT/PEDOT: PSS composite fibres. Next, the fibres were removed
1243 from the methanol bath and dried on a hotplate at 180°C for 10 min. Then, either the *p*- or *n*-type
1244 power factor of the composite fibres was improved by being immersed in hydrazine solutions or PEI
1245 infiltration, respectively. Finally, the authors fabricated a TEG by sewing 12 pairs of *p*- and *n*-type
1246 fibres (with 15 mm length) onto a PDMS substrate, see Fig. 43. While the legs were sewn in a zigzag
1247 pattern, they were electrically connected together with a silver paste. The results revealed that the TEG
1248 generated the maximum output power of 0.015 $\mu\text{W}/\text{cm}^2$ at 10°C temperature difference.

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1250 Fig. 43. Photo of the organic fibre-based TE generator sewn onto a PDMS substrate [159].
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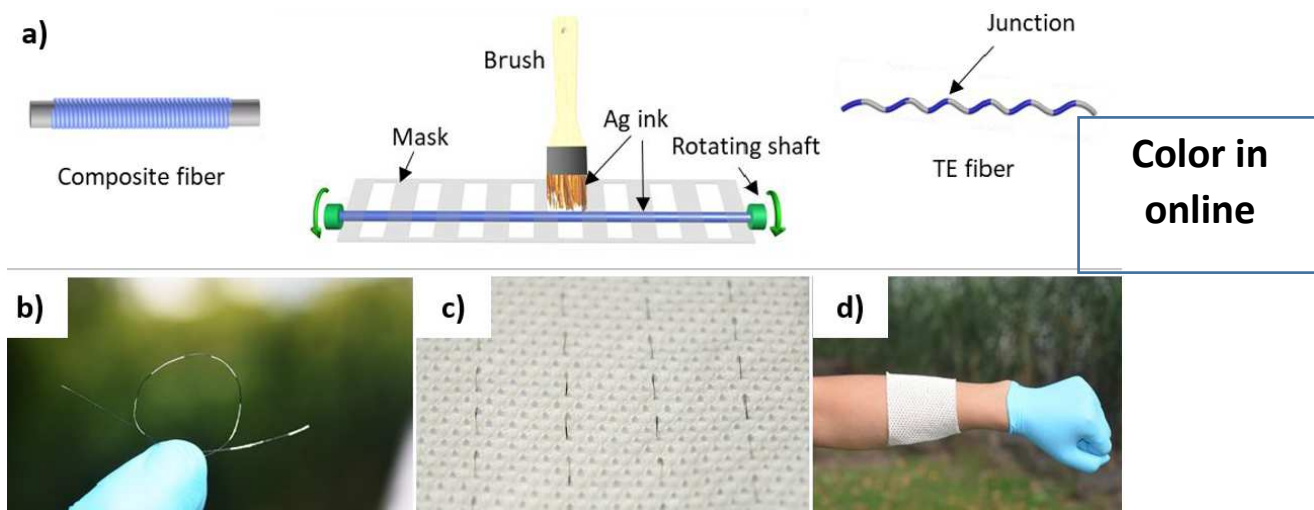
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1253 5.3. Hybrid TE materials

1254

1255 Hybrid TE materials benefit from both the low thermal conductivity and flexibility of TE polymers
1256 and the high power factor of inorganic TE materials [160]. Moreover, the combination of organic and
1257 inorganic materials arises interfacial transport properties in the resultant hybrid TE material, resulting
1258 in the enhancement of the $Z\bar{T}$ value [161]. Thus, Xu et al. [162] combined PEDOT: PSS and tellurium
1259 (Te) nanowires together to take advantage from both the high flexibility and low thermal conductivity
1260 of the former and the high Seebeck coefficient of the latter. Precisely, the authors generated *p*-type

1261 PEDOT: PSS/ Te fibres by a wet-spinning process to develop a TE textile. Accordingly, tellurium (Te)
 1262 nanowires were dispersed into deionized water followed by adding PEG and PEDOT: PSS pellets to
 1263 it, respectively. Then, the dispersions were extruded in the coagulation bath (i.e. Isopropanol solution).
 1264 The resultant fibre was collected from the bath and placed in a sealed device containing 1 mL of
 1265 ethylene glycol and dried at 120°C for 1 hr. Next, the surface of the *p*-type PEDOT: PSS/ Te fibre was
 1266 periodically brushed at equal intervals (i.e. 6.5 mm) with conductive silver paste as electrodes, see
 1267 Fig.44. Finally, the as-prepared TE fibre was sewn onto a polymer textile and worn on the arm. The
 1268 authors revealed that the highest output power of the developed TEG at ΔT of 41°C was 197.9 nW.



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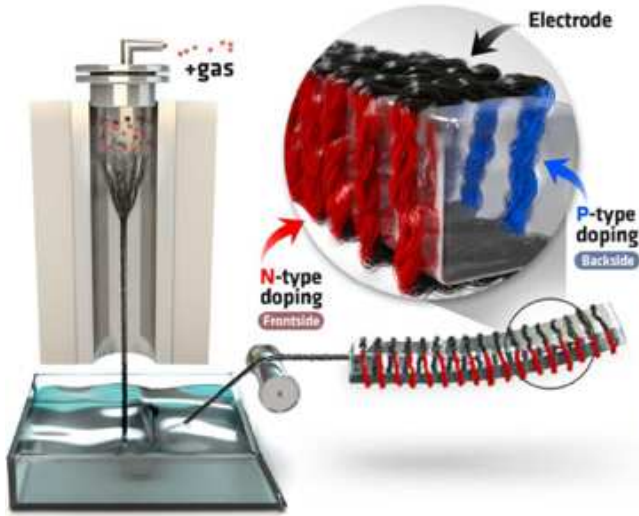
1269
 1270 **Fig. 44.** a) Schematic illustration of the fabrication processes of the TE fibres; b-d) Photos of TE fibre, TE fabric, and the
 1271 application demo [162].

1272
 1273 Choi et al. [163] considered a 3D flexible structure to not only hold the TE yarn in the 3D space, but
 1274 also use it as the supporting unit to produce the TE yarn. To specify, initially a carbon nanotube yarn
 1275 was wound around a PDMS block. Then, one side of the block was p-doped by being immersed into
 1276 FeCl₃ ethanol solution for 30 min followed by being well dried in the ambient condition, as Fig. 45
 1277 illustrates. For the n- doping, the opposite side of the PDMS block was doped with polyethyleneimine
 1278 ethanol solution for 30 min. Therefore, the two opposite un-doped sides of the PDMS block served as
 1279 electrodes. The results demonstrated that at ΔT of 65°C, the output power of the TEG increased from
 1280 1.4 μ W to 4 μ W when the number of thermocouples raised from 60 to 240. Table 4 illustrates the
 1281 summary of the reviewed literatures on wearable TEGs consisting yarn-shaped legs.

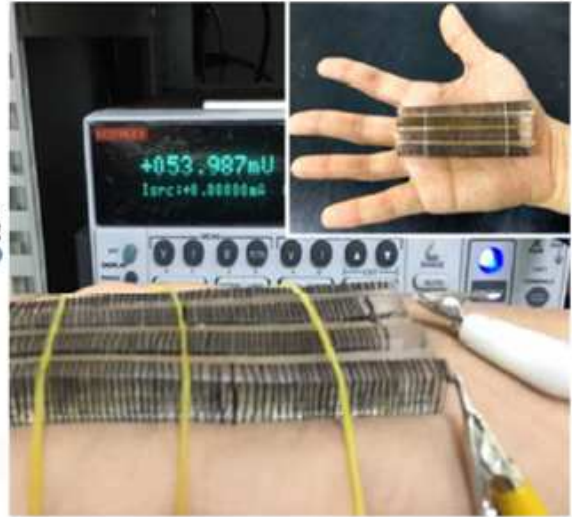
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Fig. 45. a) Schematic illustration of the doping process; b) Photo of the fabricated TEG [163].

Table 4. Summary of the recently developed wearable TEGs comprising yarn-shaped thermocouples.

<i>P</i> -type	<i>N</i> -type	Number	Electrode	Substrate	ΔT (°C)	Output	Ref
<i>Inorganic</i>							
Sb ₂ Te ₃	Bi ₂ Te ₃	30	Gold	Knitting into a textile	20	8 μ W /cm ²	[140]
SnSe	n/a	6	Copper wire	Weaving into a fabric	10	30mV	[141]
<i>Organic</i>							
PEDOT: PSS coated CNT yarn	PEI/ethanol coated CNT yarn	966	Pristine CNT yarn	Weaving into a fabric	7	0.14 μ W/cm ²	[64]
NWPU/PEDOT: PSS/MWCNT coated polyester yarn	NWPU/ MWCNT coated polyester yarn	10	Silver paint	Embroidered into the lock-nit surface of a spacer fabric	66	0.21 nW/cm ²	[142]
PEDOT:PSS coated silk thread	Silver-plated polyamide embroidery thread	8	Silver paste	Stitching through 9 layers of felted wool fabric	65	0.04 μ W/cm ²	[143]
P3HT coated cotton thread	Ag pasted P3HT/ cotton thread	13	n/a	Sewing into a fabric	50	1.15 μ W	[144]
CNT/PEG	1-butyl-3-methylimidazolium hexafluorophosphate doped CNT/PEG	-	Silver paste	Sewing into a felt fabric	15	0.125 nW/cm ²	[145]
Graphene fibres	PEIE coated graphene fibres	20	n/a	Weaving into PDMS substrate	10	1.3 pW	[146]
PEDOT:PSS doped carbon nanotube fibres	Oleamine solution sprayed carbon nanotube fibres	15	Carbon nanotube fibres	Wrapping with acrylic fibres	44	7 μ W /cm ²	[147]
PEDOT-Cl printed cotton fabric	Carbon fibres	2	Silver-coated nylon	Printing and sewing onto a commercial cotton fabric	30	0.4 nW/cm ²	[148]
PEDOT:PSS coated cotton fabric	Polyethylenimine treated carbon nanotube	8	Silver paste	Connecting with silver paste	10	10 μ W	[153]
PEDOT: PSS dyed silk yarns	PVP- MWNTs- SIS coated PET yarns	38	Conducting carbon-based paste	Sewing into a fabric	80	7.1 nW	[154]
Nickle coated carbon fibre	Carbon fibres	105	n/a	Stitching onto polyester fabric	2	0.041 nW/cm ²	[155]
PEDOT: PSS fibres	nickel wires	5	n/a	Sewing into a fabric	3.6	0.72 mV	[158]
Hydrazine immersed	PEI infiltrated CNT/PEDOT: PSS composite fibre	12	Silver paste	Sewing into PDMS substrate	10	0.015 μ W /cm ²	[159]

CNT/PEDOT: PSS composite fibre							
Hybrid							
PEDOT: PSS/ tellurium nanowires	n/a	-	Silver paste	Sewing into a polymer matrix	41	197.9 nW	[162]
FeCl ₃ ethanol doped carbon nanotube yarn	Polyethyleneimine ethanol doped carbon nanotube yarn	240	Un-doped carbon nanotube yarn	Wounding around a PDMS block	65	4 μW	[163]

1289

1290 6. Challenges and future trends

1291 As a relatively new research field, the development of wearable TEGs is confronted with several
 1292 challenges that can be classified into passive and active categories. Regarding the passive challenges,
 1293 the output voltage/power of wearable TEGs depend on several environmental factors that are variable
 1294 and out of the control of the designers, such as air temperature, air velocity, air humidity, and skin
 1295 temperature. Notably, human body sustains its basic functions by maintaining a stable internal
 1296 temperature. Thus, the brain regulates the body temperature by shivering, sweating, vasodilation, and
 1297 vasoconstriction to function within a narrow range (3°C to 4°C) from 36.8°C [164]. Based on Fig. 1,
 1298 the temperature differences between different body parts and the ambient environment are no more
 1299 than 20°C. As a result, the expected energy from wearable TEGs is in the range of between μW/cm²
 1300 and nW/cm². Therefore, the challenges for practical application of wearable TEGs at such a low
 1301 temperature difference are related to the active categories, meaning the design of the TEGs. In fact,
 1302 the key challenge is addressing the following active challenges/requirements simultaneously.

1303 First and foremost, the most fundamental requirement according to Equations 2 and 4 is developing
 1304 air-stable and high performance TE materials at room temperatures. Second, the structure of wearable
 1305 TEGs must efficiently capture the perpendicular temperature gradient between the skin and the ambient
 1306 to provide high power density. Third, wearable TEGs are required to be highly flexible to enable
 1307 sufficient thermal contact with curved surfaces of the body and avoid cumbersome for the wearers. To
 1308 fulfil the flexibility requirement, the promising approach is applying organic TE materials to fabricate
 1309 2D film- or yarn-shaped TE legs. However, this solution is in conflict with the first and second
 1310 requirements, namely; applying high performance TE materials and capturing the cross-plane
 1311 temperature gradient. This is mainly because inorganic TE materials are more efficient than the organic
 1312 ones, but due to their rigidity they are not suitable for producing 2D film- or yarn-shaped TE legs. In
 1313 addition, the 2D TE legs mostly harvest the low in-plane temperature gradient instead of the greater

1314 cross-plane one. Although it is possible to fabricate the 2D legs onto a 3D substrate, but this may cause
1315 the challenges of parasitic loss of temperature difference or offsetting the flexibility of the 2D legs.

1316 Therefore, fulfilling these three incompatible requirements at the same time is the current challenge of
1317 researchers in this field. In addition, since wearable TEGs are subject to the low temperature
1318 differences and the consequent low properties of TE materials, they cannot always fulfil the power
1319 requirement of the electronic devices. To overcome this issue, hybrid energy harvesting technology is
1320 becoming an emerging consensus. This approach benefits from both scavenging energy from multiple
1321 sources and converting energy into electricity by multiple types of transduction mechanisms [165].
1322 Accordingly, wearable TEGs can be coupled with either triboelectric generators [166], piezoelectric
1323 generators [167], or a light-to-thermal conversion layer [168] to fulfil the power requirement at
1324 different levels.

1325 Addressing the requirements of flexibility and cross-plane design at the same time, the future trend
1326 would be toward fabricating the 2D legs onto a hollow and flexible 3D supporting structure, such as
1327 Y-type [169], corrugated [170], foldable origami [171], and spring-shaped [172] structures. It should
1328 be added that so far the flexible heatsinks developed for wearable TEGs are mostly composed of
1329 heavyweight materials (e.g. copper sheets, hydrogel layers, etc.). Thus, there is a tremendous
1330 opportunity to replace the 3D heavyweight heatsinks with either hollow and flexible 3D heatsinks or
1331 2D heatsinks having higher thermal conductivities. Moving toward 2D heatsinks, it is possible to apply
1332 them for the film- and yarn-shaped legs as well. Last but not least, wearable TEGs are considered as a
1333 renewable alternative for fossil-fuel based power generations. Although a few studies carried out life
1334 cycle impact assessment of TE materials [66] and TEGs [173-174] from a specific perspective, but this
1335 part is heavily neglected in the existing research on wearable TEGs. Therefore, as part of the
1336 sustainability criteria for future technologies, the environmental performance of wearable TEGs
1337 requires careful attention.

1338 7. Conclusions

1339 This review provides an overview of different materials and configurations of the TE legs in wearable
1340 TEGs to pave the way for comparing the output voltages/powers of the resultant TEGs with each other.
1341 Accordingly, it demonstrates both the procedure of fabricating different configurations with different
1342 TE materials and the resultant output voltages/powers of the wearable TEGs. Among different types

1343 of TE materials, the inorganic ones specifically Bi_2Te_3 and its alloys are the most popular candidate to
1344 generate the ingot-shaped thermocouples. It has been attempted to improve the flexibility of the ingot-
1345 shaped legs by embedding them in a polymer-based flexible substrate. However, fully coating the legs
1346 with the polymer-based substrate negatively affects the output power of the wearable TEGs. To
1347 overcome this issue, high thermal conductivity additives can be added to the flexible substrates around
1348 the junctions. In addition, several heatsinks have been developed to further increase the output power
1349 of the ingot-shaped legs. However, since these heatsinks are bulky, they add to the weight of the
1350 wearable TEGs and inevitably cause cumbersome for the wearers. To solve these problems, the
1351 researchers switched their focus from the ingot-shaped legs to the film-shaped one. Notably, the
1352 contribution of all the three types of TE materials to the preparation of film-shaped legs is the same.
1353 Comparing the output power of the inorganic ingot-shaped thermocouples with that of the inorganic
1354 film-shaped ones, obviously the former is superior to the latter. To specify, to generate a specific
1355 amount of output power, the inorganic film-shaped legs require higher temperature gradient and
1356 number of thermocouples than the inorganic ingot-shaped ones. To solve this issue, some researchers
1357 attempted to replace the 2D underlying substrate of the film-shaped legs (mostly a polyimide-based
1358 substrate) with a 3D one such as a PDMS cable or block. Based on these attempts, yarn-shaped TE
1359 legs have been developed, which could be easily sewn onto fabrics and be worn. Based on the reviewed
1360 papers, the organic TE materials are the most dominant TE materials utilized for developing the yarn-
1361 shaped legs. Comparing the organic film-shaped thermocouples with the yarn-shaped ones, the former
1362 provides greater output power due to its larger contact area with the heat source (the skin). Therefore,
1363 further improvement of the wearable TEG requires addressing the flexibility and weight of the
1364 heatsinks, developing more efficient organic and hybrid TE materials, and increasing the contact area
1365 between the yarn-shaped legs and the skin.

1366

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