A Wavy-Structured Highly Stretchable Thermoelectric Generator with Stable Energy Output and Self-Rescuing Capability

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Thermoelectric generators (TEGs) demonstrate great potential for flexible and wearable electronics due to the direct electrical energy harvested from waste heat. Good wearability requires high mechanical flexibility and preferable stretchability, while current TEGs are primarily developed with rigid or non-stretchable components, which do not conform well to human skin or accommodate human motions, thus hindering further applications. Herein, a wavy architecture was proposed to fabricate stretchable TEGs, wherein a stretchable and self-healable hydrogel was employed as a device substrate, and intrinsically flexible, high-performance thermoelectric (TE) films were attached to form wavy morphologies. The wavy-structured TEG could be stretched readily to 300%; in the meantime. It could sustain a stable energy output able to retain more than 90% TE performance, thus, outperforming most of the reported state-of-the-art TE materials and TEGs. It also demonstrated a desired device-level self-rescuing capability that originated from the effective self-healing of the hydrogel and the wavy structure of TE legs, thereby providing constant energy supply upon injuries or damages. This work offers a promising approach to the design of next-generation stretchable and wearable TEGs.

Keywords: flexible devices, thermoelectrics, energy conversion, stretchable, wearable

Introduction

With the rapid development of wearable electronics for the applications of motion monitoring, health monitoring, the internet of things, virtual reality, and sensory enhancement, the need for flexible and wearable energy supply systems has aroused considerable interest in recent years.1–4 The human body, as a constant heat source, can potentially act as an in situ energy base if the waste body heat can be conveniently collected and efficiently converted into electrical power.5 As a green and sustainable energy technique, thermoelectric generators (TEGs) can convert waste heat into electricity directly without the involvement of moving parts or emission of waste
Harvesting energy from the human body via TEGs, thus, represents a promising direction in designing self-powered wearable electronics, especially when the cutting-edge technologies have significantly reduced their power requirement, suggesting a bright future for the practical applications of thermoelectric energy harvesting.\(^\text{29,30,32}\) Whereas, existing TEGs are primarily fabricated using rigid or non-stretchable components, which restrains their capability to conform to human skin or accommodate human motions.\(^\text{31}\) To meet the requirement of wearability, TEGs must possess high mechanical flexibility and preferable stretchability, and in the meantime, be able to provide consistent energy output, even upon encountering mechanical damages.

Recently, a few efforts have been devoted to developing flexible and stretchable TEGs for wearable applications. The design approaches of these TEGs can essentially be summarized into three categories: (1) The first is to simply integrate rigid TE legs into extendable matrices or to attach flexible TE films onto stretchable substrates.\(^\text{16,17}\) These TEGs inherited conventional device design routes and exhibited moderate stretchability (<100%). Due to the mismatch in the mechanical properties between the TE materials and the stretchable matrices/substrates, for these TEGs, performance deterioration is usually inevitable upon stretching. (2) The second involves the synthesis of intrinsically extendable TE materials,\(^\text{18–21}\) or the construction of intrinsically stretchable TE composites by introducing elastic matrixes.\(^\text{22–26}\) Despite the relatively stable Seebeck coefficient, the former approach could hardly sustain constant electrical conductivity during stretching, and the stretchability is substantially limited by the intrinsic mechanical properties of the TE materials (usually <50%). The latter approach harnessed the elasticity of the matrices; thus, it could achieve higher stretchability (>100%) for the resultant TE composites, but their electrical conductivity decayed drastically with stretching. (3) The third is to realize device-level stretchability via rational device structure design. This approach resulted in high stretchability (60–200%) with stable TE performance.\(^\text{27–32}\) For example, Jeong et al.\(^\text{31}\) reported using a liquid alloy as connecting wires for Bi\(_2\)Te\(_3\) TE legs, and Yang et al.\(^\text{27}\) exploited wavy serpentine interconnected to integrate p–n units, both of which resulted in stretchable TEGs. Besides, researchers have conceived various TEGs based on novel helical architectures, imparted the intrinsically rigid TE materials with device-level flexibility and stretchability.\(^\text{29,30,32}\) Furthermore, Sun et al.\(^\text{28}\) developed TE fabrics woven out of TE fibers to ensure high compatibility with body movements, and the designed interlocked structure enabled good stretchability (80%) and sustained energy supply. These reported stretchable TEGs usually require a delicate or complex device fabrication process; they lack the device-level self-rescuing capability to recover from external mechanical damages, implying that a stable energy supply would be easily interrupted once device damage is induced.

The desired features of wearable TEGs include high TE performance at near room temperature, compliant mechanical flexibility for curved or irregular skins, high stretchability to accommodate strains induced by body motions and self-rescuing capability to heal mechanical damages in practical operation. Hydrogels are hydrophilic polymer networks swollen with considerable amounts of water, stretchable and conformal to human skins and can be designed with self-healing ability through dynamic bonds.\(^\text{33–35}\) In this study, a flexible TEG for wearable applications was fabricated by employing a micro-wavy architecture combining high-performance, intrinsically non-stretchable TE films with an elastic hydrogel substrate. The hydrogel possessed a covalently cross-linked network with molecular chains interacting with each other via multiple dynamic hydrogen bonding, thus, giving high stretchability and self-healing capability. The designed micro-wavy architecture translated the high stretchability and self-healing capability from the hydrogel substrate into the device, affording a highly stretchable TEG with desired self-rescuing capacity. The TEG developed in this way stretched readily to 300% without deteriorating its TE performance and could readily remedy injuries or damages via self-healing of the hydrogel and alignment of the wavy structure. This work provides a simple, facile, and effective approach for developing next-generation stretchable and wearable TEGs.

**Experimental Methods**

**Materials**

Acrylic acid (AA), N,N,N-methylenebisacrylamide (MBAA), and polyethyleneimine (PEI) were purchased from Shanghai Aladdin Reagent Chemistry Co., Ltd. (Shanghai, China). Ammonia persulfate (APS) was provided by Shanghai Macklin Biochemical Technology Co., Ltd. (Shanghai, China). Carbon nanotube papers (CNTPs) were purchased from Suzhou Hengqiu Nanotech Co., Ltd. (Suzhou, Jiangsu, China). All the chemicals were used as received without any purification or treatment. Deionized water was employed in all experiments.

**Preparation of the hydrogel, p-type TE legs, and n-type TE legs**

Precisely 14.7 g AA monomer was first dissolved in 40 mL deionized water, and then 7.4 mg MBAA and 22.3 mg APS were added as covalent cross-linker and initiator, respectively, followed by stirring for 1 h at room temperature. The reaction mixture obtained was degassed by ultrasonic treatment and then poured into glass moulds, followed by polymerization at 65 °C in an oven overnight. The p-type TE legs were directly obtained using pristine...
CNTP films. The n-type TE legs were prepared by doping the pristine CNTP films with PEI. Specifically, the pristine CNTP films were immersed in 1 wt% aqueous PEI solution for 2 h and were then transferred to polytetrafluoroethylene plate without rinse. These films were further heated and dried in a vacuum oven at 80 °C for 2 h, affording the n-type PEI-CNTP films.

Characterizations

The micromorphology of each sample was collected on a Zeiss EVO MA15 (Oberkochen, Germany) scanning electron microscopy (SEM) with an acceleration voltage of 10 kV. Attenuated total reflection Fourier transform infrared (ATR-FTIR) spectroscopy (PerkinElmer Spectrum 3, Waltham, Massachusetts, USA) was used to analyze the structure and components of the samples. A dry hydrogel was cut into a small piece and scanned in the wavenumber range of 4000–450 cm⁻¹ with 64 scans and a nominal resolution of 4 cm⁻¹ at room temperature. The Raman spectra of each sample were collected on a Raman spectrometer (Horiba LabRAM HR Evolution, Kyoto, Japan) with a laser at an excitation wavelength of 532 nm. The surface chemical composition of the CNTP samples was analyzed on a Thermo ESCALAB 250XI X-ray photoelectron spectroscopy (XPS) (Waltham, Massachusetts, USA). The X-ray diffraction (XRD) patterns were obtained using a Rigaku MiniFlex 600 (Tokyo, Japan) instrument. The electrical conductivity and the Seebeck coefficient were measured on a commercial Film Thermoelectric Parameter Test System (Joule Yacht MRS-3; Joule Yacht Technology Co., Ltd., Wuhan, China). The stretchable TEG was fabricated by attaching the TE legs onto the hydrogel substrate, where copper foils and copper wires were employed as electrodes and connecting leads, respectively. The hydrogel substrate was pre-stretched at 300% strain before the attachment of TE legs. The TE performance of the TEGs was evaluated by a self-established apparatus consisting of a cold plate, a hot plate, a temperature control system, and a Keithley 2700 Data Acquisition (Solon, Ohio, USA). The output voltage (V) and output current (I) were recorded with varying loading resistance. The output power (P) was calculated according to the formula \( P = I \times V \).

Results and Discussion

The hydrogel serving as a conformal substrate was the key to realizing high stretchability and preferable self-rescuing capability for the desired TEG. As illustrated in Figure 1a, the hydrogel consisted of a covalently cross-linked polyacrylic acid (PAA) network swollen with a large amount of water (73% by weight), imparting good stretchability without fragmentation, and the multiple dynamic inter- and intramolecular hydrogen bonds enabled an effective self-healing capability (Figure 1b). It should be noted that high water content was required to achieve high softness and flexibility, supporting guaranteed good adaptivity to human skin. However, water molecules could weaken the strength of the inter- and intramolecular hydrogen bonds, lowering the self-healing efficiency. The hydrogel synthesis involved polymerization of AA monomers in glass moulds, where APS and MBAA were utilized as an initiator and cross-linking agent, respectively (Figure 1c). The synthesis route is further illustrated in Supporting Information Figure S1. The resultant flexible and stretchable hydrogel could be readily tailored into various shapes and sizes; thus, well satisfying the dimension requirement of the subsequent device fabrication.

As revealed by the SEM images in Figures 2a and 2b, the hydrogel possessed a highly porous honeycomb structure due to the formation of an interconnected polymer network and the swelling with water, which also signified its high stretchability. FTIR spectrum confirmed the structural information of PAA. As shown in Figure 2c, the peak at 794 cm⁻¹ should be designated as the CH₂ rocking mode of PAA, and the peak at 1165 cm⁻¹ is ascribed to the C=O stretching mode. A sharp peak at 1694 cm⁻¹ was attributed to the C=O stretching mode of the carboxylic group in PAA. The broad bands at 2861 and 2923 cm⁻¹ originated from the sp³ C–H stretching mode. The mechanical properties of the hydrogel were evaluated on a tensile tester. According to the stress–strain curve presented in Figure 2d, the hydrogel showed an elastic deformation upon stretching up to ~1300%. The high stretchability was further demonstrated, as shown in the insets of Figure 2d, and the corresponding mechanical properties are listed in Supporting Information Figure S2. Besides the impressive stretchability, the hydrogel also possessed a desirable seal-healing feature. As shown in Figure 2e, a thin piece of hydrogel was cut into two halves and was then healed within minutes. The self-healing process was proceeded by resting the hydrogel in an ambient environment, and the cut-wound recombined without the need for external stimuli. Undoubtedly, longer self-healing time further increased the bonding strength at the wound. The carbonyl groups in PAA provided abundant and stable hydrogen bonding, and the facile self-healing capability originated from the multiple dynamic hydrogen bonds existing within and between the PAA molecular chains, which were reversible interactions and could be rebuilt readily. Figure 2f demonstrates the strong bonding energy at the wound, suggesting an effective self-healing.
The hydrogel only contributes to the device-level stretchability and self-rescuing capability, without providing any actual TE performance. Therefore, CNTP was employed as the active TE material for the TEG design. Specifically, the p-type TE legs were directly obtained using pristine CNTP films, and the n-type TE legs were prepared by doping the pristine CNTP with a reducing agent. PEI bears a high density of amine groups and thus, was chosen as an efficient dopant for CNTP. The p-type nature of the pristine CNTP was attributed to the adsorbed surficial oxygen-containing groups on the CNTP surface that caused a nonnegligible hole-doping, and the n-type nature of the PEI-CNTP resulted from an increased electron concentration, facilitating facile electron transfer from PEI to CNTP. SEM images revealed an interconnected carbon nanotube network within the CNTP; the nanotubes were covered and wrapped with a polymer “skin” after PEI doping (Figures 3a and 3b and Supporting Information Figure S3). Raman spectra showed the characteristic D band, G band, and 2D band for both samples (Figure 3c), corresponding to the defect-activated hexagon-breathing mode, C–C in-plane bond stretching mode, and a vibrational mode, characterized by breathing of the hexagonal lattice of graphene, respectively. XRD patterns of the two samples are shown in Supporting Information Figure S4. XPS analysis further revealed the structural information of the CNTP after PEI doping, where N1s spectrum was identified for PEI-CNTP, and the peak for C–N bonding emerged in its C1s spectrum (Figures 3d–3f). The O1s spectra of both samples are shown in Supporting Information Figure S5. The mechanical properties of the TE films were also evaluated using a tensile tester. According to the stress–strain curves, the pristine CNTP exhibited a high elastic modulus of 2767.4 MPa and high breaking stress of 98.3 MPa; notably, these values were reduced to 836.0 and 50.1 MPa, respectively, after PEI doping. In the meantime, the elongation at break obviously increased from 14.3% to 22.9% (Figures 3g and 3h). Thus, the doping of PEI introduced soft polymer into the pristine CNTP network, resulting in softer n-type TE films. The average thickness of the CNTP was ~25 μm, and this number slightly increased to 28.5 μm after PEI doping. Both the CNTP and the PEI-CNTP sustained high flexibility and could be folded, twisted, and knotted into various shapes, as demonstrated in Figure 3i.

As presented in Figure 4a and Supporting Information Figure S6, the pristine CNTP exhibited a Seebeck coefficient of 60.8 μV K⁻¹ and electrical conductivity of 877.5 S cm⁻¹ at room temperature, leading to a power factor (PF) of 324.87 μW m⁻¹ K⁻². After PEI doping, the p-type CNTP was converted successfully into n-type, demonstrating a

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**Figure 1** | (a) Schematic illustration of the hydrogel structure. (b) Schematic illustration of the self-healing nature of the hydrogel via multiple dynamic hydrogen bonds. (c) Schematic illustration showing the fabrication process of the hydrogel.
negative Seebeck coefficient of $-59.9\ \mu V\ K^{-1}$. The electrical conductivity was also improved to $1056.3\ S\ cm^{-1}$, attributable to the reducing effect of PEI that pushed out the surficial oxygen. As a result, the PF value of the n-type PEI-CNTP was calculated to be $378.97\ \mu Wm^{-1}K^{-2}$, comparable with that of the n-type CNTP, suggesting a good match between the p- and n-type materials in thermoelectric performance. Note that the doping time significantly affected the TE performance of the resultant PEI-CNTP, with a saturated doping time estimated as 2 h in an 80 °C oven in a vacuum (Supporting Information Figure S7). Considering reoxidization by air in an ambient environment, the air stability of the n-type PEI-CNTP was also evaluated. As shown in Figure 4b, the n-type PEI-CNTP sustained an almost constant Seebeck coefficient during its 7-day exposure under ambient conditions, during which the electrical conductivity was slightly decreased with exposure time. After the 7-day exposure in an ambient environment, the morphology and structure of the n-type PEI-CNTP remained almost unchanged (Supporting Information Figure S8), and 88.4% of its initial PF value was retained, which is acceptable considering that extra packaging is usually adopted under these conditions; thus, most of the air and moisture could be isolated, with an anticipation of better stability. Figures 4c and 4d show the TE performance of the pristine CNTP and PEI-CNTP as a function of temperature (measured in vacuum). For the p-type CNTP, the Seebeck coefficient slightly increased at first and then gradually decreased, and the electrical conductivity showed a decreasing trend with increased temperature, displaying typical metal-like behavior. Thus, the resultant PF value decreased with increasing temperature. For the n-type PEI-CNTP, the Seebeck coefficient first decreased and then gradually increased, and the electrical conductivity increased in the beginning, possibly due to the further reduction of carbon nanotubes at elevated temperatures in a vacuum until the effect of metal-like behavior surpassed the marked decline. As a result, the PF value of the PEI-CNTP increased with increasing temperature.

Given the high TE performance of the CNTP and PEI-CNTP films, a flexible TEG was subsequently fabricated employing a thin layer of PAA hydrogel as a substrate. As illustrated in Figure 5a, the hydrogel layer was prestretched before attaching the CNTP and PEI-CNTP TE legs; it was then released to its original length, affording a wavy-structured device with reversible stretchability (Supporting Information Figure S9). Due to the sticky nature of the PAA hydrogel, good contact between the TE legs and the hydrogel could be ensured without an extra binder (Supporting Information Figure S10). The TE leg adopted a passive wavy structure, as shown in the
This design route well translated the stretchability and self-healing feature of the hydrogel into the device, resulting in a flexible TEG with high stretchability and self-rescuing capability. Notably, the width of hydrogel substrate would dramatically shrink upon extensive stretching; hence, it could not provide enough space to accommodate the TE leg array. Therefore, to ensure sufficient space for the array of TE legs, the stretchability of the TE device was set at 300%. As shown in Figure 5b and Supporting Information Figure S12, the as-fabricated TEGs demonstrated linearly increased open-circuit voltage with increasing temperature or the number of p–n couples, suggesting good scalability. Besides, due to the nondestructive wavy structure, the TEG could be stretched readily to 300% strain without deteriorating the electrical conductivity and the Seebeck coefficient of the TE legs (Supporting Information Figures S13 and S14), leading to stable PF values against various mechanical strains (Supporting Information Figure S15). The resultant open-circuit voltage was also well maintained during the 300% stretching period (Supporting Information Figure S16), with multiple stretching cycles (Figure 5c) without notable fluctuations. This is rather advantageous
compared with the TEGs using intrinsically stretchable TE materials, as those TEGs were usually much less stretchable and could hardly maintain a stable TE performance when being stretched to various strains. It was verified that the water contained in the hydrogel (or the ambient humidity) did not affect the TE performance, as the TE properties of the CNTP and PEI-CNTP originated from the transport of holes and electrons, and no Soret effect contributed to the overall TE performance. The output energy data, as a function of current, are presented in Figure 5d, which revealed that, for a small temperature difference of 15 K, a high output voltage of 4.15 mV and high output power of 172.9 nW were achieved with only three pairs of p-n couples. Such energy output could well meet the requirement of many sensors and transmitters with low power consumption. The output energy data of the TEGs consisting of 1 and 5 p-n couples are listed in Supporting Information Figure S17, and the corresponding output voltage and output current as a function of loading resistance are presented in Supporting Information Figures S18 and S19, respectively. As shown in Supporting Information Figure S20, the peak output power values for the TEGs with 1, 3, and 5 p-n couples were recorded at the loading resistance of ~14.5, 25.8, and 60.0 Ω, respectively, which well matched the measured internal resistances of the corresponding TEGs (12.3, 20.8, and 53.8 Ω). That is to say, the output power $P_{\text{power}}$ reached its maximum when the loading resistance $R_L$ equaled the internal resistance $R_s$, as supported by the equation $P_{\text{power}} = \frac{V_o c^2}{2R_s}$, where $V_{oc}$ denotes the output voltage. The difference between the fitted and the measured internal resistance values should be mainly attributable to the inevitable contact resistance brought about by connecting wires and electrodes.

As an electrical conductivity, the Seebeck coefficient and the open-circuit voltage remained almost constant under various strains; the stable energy output of the stretchable TEG was further verified and demonstrated. As shown in Figures 5e and 5f, when stretched to 100% strain and 300% strain at the fixed temperature difference of 15 K, the output energy of the TEG was still well sustained. The output voltage and current were only slightly affected, which is within the reasonable fluctuations possibly induced by the applied stress, causing a slight variation in device resistance, as well as...
temperature gradient distribution. Besides the stable TE performance against high stretchability, a desired self-rescuing function benefited from the self-healing merit of the PAA hydrogel was also anticipated. The TE performance retention of the stretchable TEG was evaluated after each operation of self-rescuing. Specifically, one TE leg of the TEG was half-cut in width and healed, denoted as the first healing. Another TE leg was totally cut off in width and then healed, denoted as the second healing. The third healing involves a total cutting off and the subsequent healing of a third TE leg. (The cutting process is illustrated in Supporting Information Figure 5)

Figure 5 | (a) Schemes, showing the design principle of the wavy-structured stretchable TEG. The yellow objects represent p-type CNTP or n-type PEI-CNTP TE legs, and the blue objects represent the hydrogel substrates. An enlarged inset shows the cross-section of the TEG, where CNTP or PEI-CNTP films are tightly attached to the hydrogel surface, forming a wavy structure that allows for reversible stretching. (b) Comparison of the open-circuit voltage of the TEGs consisting of various p–n couples as a function of temperature. (c) The output voltage variation of the TEG consisting of 1 p–n couple with multiple stretching cycles under the temperature difference of 10 K. (d) Output voltage and output power of the TEG consisting of 3 p–n couples as a function of output current at the temperature difference of 15 K. (e) Output voltage variation of the TEG consisting of 3 p–n couples under different stretching ratios at the temperature difference of 15 K. (f) Output power variation of the TEG consisting of 3 p–n couples under different stretching ratios at the temperature difference of 15 K. (g) Energy output of the TEG consisting of 3 p–n couples before and after self-rescuing at the temperature difference of 15 K. (h) Schematic representation of the self-rescuing mechanism of the TEG. (i) Comparison of the stretchability and the TE performance retention with literature-reported TE materials and TEGs.
Figure S21.) As shown in Figure 5g, each self-rescuing operation resulted in reduced output energy. After the third healing, the output current, voltage, and power retention of TEG were 80.1%, 80.9%, and 64.8%, respectively, compared with its initial values. The underlying self-rescuing mechanism of the TEG is illustrated in Figure 5h: After cutting, both the hydrogel substrate and the TE leg were left broken. On the one hand, due to the effective self-healing feature, the hydrogel could be repaired readily. On the other hand, benefitting from the wavy structure, the broken TE leg possessed sufficient margin to reattach to the hydrogel surface and reconnect with each other. As a result, the TEG was self-rescued; thus, the malfunction was avoided. Although the energy output could not be fully restored after self-rescuing, the TEG demonstrated a capability of consistently working to meet any urgent need. Also, it is worth noting that both the construction of the self-healable hydrogel and the rational wavy-structured device design are crucial to realizing device-level stretchability and self-rescuing capability. The stretchability and the corresponding TE performance retention during stretching were compared with state-of-the-art stretchable TE materials and TEGs. As shown in Figure S5i, the TEG developed in this work yielded a 300% stretchability with over 90% TE performance retention, which outperformed most of the reported results in the literature. The corresponding metrics of these stretchable TEGs are listed in Supporting Information Table S1.

**Conclusion**

A highly stretchable TEG with stable energy output and self-rescuing capability was proposed via the rational design of a micro-wavy architecture. The device architecture employed stretchable and self-healable PAA hydrogel as substrate and intrinsically flexible but non-stretchable CNTP and PEI-CNTP films as TE legs. Benefiting from the wavy structure, the as-fabricated TEG well inherited high stretchability and demonstrated stable energy output under various strains: It could well sustain more than 90% of its initial energy output at an extensive stretching of 300%, which outperformed most of the state-of-the-art stretchable TE materials and TEGs. Besides its high stretchability and stable TE performance, the TEG could also realize efficient device-level self-rescuing through self-healing of the hydrogel electrolyte and reattachment and reconnection of the wavy TE legs, resulting in continuous energy supply without malfunction when subjected to mechanical injuries or damages. The stable energy output under large strains and the self-rescuing capability are both desirable features for flexible and wearable electronics, suggesting a high potential of the TEG in wearable applications.

**Supporting Information**

Supporting Information is available and includes additional illustrations, supplementary mechanical property data, thermoelectric testing results, SEM images, XRD patterns, XPS spectra, Raman spectra, and the metrics for comparing various stretchable TEGs.

**Conflict of Interest**

There is no conflict of interest to report.

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**References**

1. Lee, G.-H.; Moon, H.; Kim, H.; Lee, G. H.; Kwon, W.; Yoo, S.; Myung, D.; Yun, S. H.; Bao, Z.; Hahn, S. K. Multifunctional Materials for Implantable and Wearable Photonic Health Devices. *Nat. Rev. Mater.* 2020, 5, 149–165.
2. Son, D.; Lee, J.; Qiao, S.; Ghaffari, R.; Kim, J.; Lee, J. E.; Song, C.; Kim, S. J.; Lee, D.; Jun, S. W.; Yang, S.; Park, M.; Shin, J.; Do, K.; Lee, M.; Kang, K.; Hwang, C. S.; Lu, N.; Hyeon, T.; Kim, D.-H. Multifunctional Wearable Devices for Diagnosis and Therapy of Movement Disorders. *Nat. Nanotechnol.* 2014, 9, 397–404.
3. Wang, B.; Thukral, A.; Xie, Z.; Liu, L.; Zhang, X.; Huang, W.; Yu, X.; Yu, C.; Marks, T. J.; Facchetti, A. Flexible and Stretchable Metal Oxide Nanofiber Networks for Multimodal and Monolithically Integrated Wearable Electronics. *Nat. Commun.* 2020, 11, 2405.
4. Moin, A.; Zhou, A.; Tahimi, A.; Menon, A.; Benatti, S.; Alexandrov, G.; Tamakloe, S.; Ting, J.; Yamamoto, N.; Khan, Y.; Burghardt, F.; Benini, L.; Arias, A. C.; Rabaey, J. M. A Wearable Biosensing System with In-Sensor Adaptive Machine Learning for Hand Gesture Recognition. *Nat. Electron.* 2021, 4, 54–63.
5. Qi, Y.; McAlpine, M. C. Nanotechnology-Enabled Flexible and Biocompatible Energy Harvesting. *Energy Environ. Sci.* 2010, 3, 1275–1285.
6. Bell, L. E. Cooling, Heating, Generating Power, and Recovering Waste Heat with Thermoelectric Systems. *Science* 2008, 327, 1457–61.
7. Vining, C. B. An Inconvenient Truth about Thermoelectrics. *Nat. Mater.* 2009, 8, 83–5.
8. Lim, W.; Lee, I.; Sylvester, D.; Blaauw, D. B. Batteryless Sub-NW Cortex-M0+ Processor with Dynamic Leakage-Suppression Logic, in 2015 IEEE International Solid-State Circuits Conference (ISSCC), Digest of Technical Papers, 2015, pp 1–3. DOI: 10.1109/ISSCC.2015.7062968.
9. Mercier, P. P.; Bandypadhyay, S.; Lysaght, A. C.; Stankovic, K. M.; Chandrasan, A. P. A Sub-nW 2.4 GHz Transmitter for Low Data-Rate Sensing Applications. *IEEE J. Solid-State Circuits* 2014, 49, 1463–1474.
10. Park, H.; Lee, D.; Park, G.; Park, S.; Khan, S.; Kim, J.; Kim, W. Energy Harvesting Using Thermoelectricity for IoT (Internet of Things) and E-Skin Sensors. *J. Phys. Energy* 2019, 1, 042001.
11. Liang, J.; Wang, T.; Gou, P.; Yang, S.; Ming, C.; Chen, H.; Song, Q.; Zhao, K.; Wei, T.-R.; Ren, D.; Sun, Y.-Y.; Shi, X.; He, J.; Chen, L. Flexible Thermoelectrics: From Silver Chalcogenides to Full-Inorganic Devices. *Energy Environ. Sci.* 2019, 12, 2983–2990.
12. Kishore, R. A.; Nozariarismarz, A.; Poudel, B.; Sanghadasa, M.; Priya, S. Ultra-High Performance Wearable Thermoelectric Coolers with Less Materials. *Nat. Commun.* 2019, 10, 1765.
13. Hong, S.; Gu, Y.; Seo, J. K.; Wang, J.; Liu, P.; Meng, Y. S.; Xu, S.; Chen, R. K. Wearable Thermoelectrics for Personalized Thermoregulation. *Sci. Adv.* 2019, 5, eaaw0536.
14. Li, A.; Fu, C.; Zhao, X.; Zhu, T. High-Performance Mg3Sb2-xbi Thermoelectrics: Progress and Perspective. *Research 2020*, 2020, 1934848.
15. Jiang, B.; Yu, Y.; Cui, J.; Liu, X.; Xie, L.; Liao, J.; Zhang, Q.; Huang, Y.; Ning, S.; Jia, B.; Zhu, B.; Bai, S.; Chen, L.; Pennycook, S. J.; He, J. High-Entropy-Stabilized Chalcogenides with High Thermoelectric Performance. *Science 2021*, 371, 830–834.
16. Zadan, M.; Malakooti, M. H.; Majidi, C. Soft and Stretchable Thermoelectric Generators Enabled by Liquid Metal Elastomer Composites. *ACS Appl. Mater. Interfaces 2020*, 12, 17921–17928.
17. Jeong, M. H.; Sanger, A.; Kang, S. B.; Jung, Y. S.; Oh, I. S.; Yoo, J. W.; Kim, G. H.; Choi, K. J. Increasing the Thermoelectric Power Factor of Solvent-Treated PEDOT-PSS Thin Films on PDms by Stretching. *J. Mater. Chem. A* 2018, 6, 15621–15629.
18. Liang, L.; Gao, C.; Chen, G.; Guo, C.-Y. Large-Area, Stretchable, Super Flexible and Mechanically Stable Thermoelectric Films of Polymer/Carbon Nanotube Composites. *J. Mater. Chem. C* 2016, 4, 526–532.
19. Ke, S.; Kim, H.; Paleti, S. H. K.; El Labban, A.; Neophytou, M.; Emwas, A.-H.; Alshareef, H. N.; Baran, D. Highly Stretchable and Air-Stable PEDOT:PSS/Ionic Liquid Composites for Efficient Organic Thermoelectrics. *Chem. Mater.* 2019, 31, 3519–3526.
20. Zhang, C.; Zhang, Q.; Zhang, D.; Wang, M.; Bo, Y.; Fan, X.; Li, F.; Liang, J.; Huang, Y.; Ma, R.; Chen, Y. Highly Stretchable Carbon Nanotubes/Polymer Thermoelectric Fibers. *Nano Lett.* 2021, 21, 1047–1055.
21. Li, H.; Mao, P.; Davis, M.; Yu, Z. PEDOT-PSS-Polyethylene Oxide Composites for Stretchable and 3D-Printed Thermoelectric Devices. *Composites Communications* 2021, 23, 100599.
22. Taroni, P. J.; Santagugiana, G.; Wan, K.; Calado, P.; Qiu, M.; Zhang, H.; Pugno, N. M.; Palma, M.; Stingelin-Stutzman, N.; Heeney, M.; Fenwick, O.; Baxendale, M.; Bilotti, E. Toward Stretchable Self-Powered Sensors Based on the Thermoelectric Response of PEDOT:PSS/Polyurethane Blends. *Adv. Funct. Mater.* 2018, 28, 1704285.
23. Jeong, Y. J.; Jung, J.; Suh, E. H.; Yun, D. J.; Oh, J. G.; Jang, J. Self-Healable and Stretchable Organic Thermoelectric Materials: Electrically Percolated Polymer Nanowires Embedded in Thermoplastic Elastomer Matrix. *Adv. Funct. Mater.* 2019, 30, 1905809.
24. Kee, S.; Haque, M. A.; Corzo, D.; Alshareef, H. N.; Baran, D. Self-Healing and Stretchable 3D-Printed Organic Thermoelectrics. *Adv. Funct. Mater.* 2019, 29, 1905426.
25. Wan, K.; Taroni, P. J.; Liu, Z.; Liu, Y.; Tu, Y.; Santagugiana, G.; Hsia, I. C.; Zhang, H.; Fenwick, O.; Krause, S.; Baxendale, M.; Schroeder, B. C.; Bilotti, E. Flexible and Stretchable Self-Powered Multi-Sensors Based on the N-Type Thermoelectric Response of Polyurethane/Na(Ni-ett)2 Composites. *Adv. Electron. Mater.* 2019, 5, 1900582.
26. Zhang, D.; Zhang, K.; Wang, Y.; Wang, Y.; Yang, Y. Thermoelectric Effect Induced Electricity in Stretchable Graphene-Polymer Nanocomposites for Ultrasensitive Self-Powered Scan Sensor System. *Nano Energy 2019*, 56, 25–32.
27. Yang, Y.; Hu, H.; Chen, Z.; Wang, Z.; Jiang, L.; Lu, G.; Li, X.; Chen, R.; Jin, J.; Kang, H.; Chen, H.; Lin, S.; Xiao, S.; Zhao, H.; Xiong, R.; Shi, J.; Zhou, Q.; Xu, S.; Chen, Y. Stretchable Nanolayered Thermoelectric Energy Harvester on Complex and Dynamic Surfaces. *Nano Lett.* 2020, 20, 4445–4453.
28. Sun, T.; Zhou, B.; Zheng, Q.; Wang, L.; Jiang, W.; Snyder, G. J. Stretchable Fabric Generates Electric Power from Woven Thermoelectric Fibers. *Nat. Commun.* 2020, 11, 572.
29. Xu, X.; Zuo, Y.; Cai, S.; Tao, X.; Zhang, Z.; Zhou, X.; He, S.; Fang, X.; Peng, H. Three-Dimensional Helical Inorganic Thermoelectric Generators and Photodetectors for Stretchable and Wearable Electronic Devices. *J. Mater. Chem. C* 2018, 6, 4866–4872.
30. Nan, K. W.; Kang, S. D.; Li, K.; Yu, K. J.; Zhu, F.; Wang, J. T.; Dunn, A. C.; Zhou, C. Q.; Xie, Z. Q.; Agne, M. T.; Wang, H. L.; Luan, H. W.; Zhang, Y. H.; Huang, Y. G.; Snyder, G. J.; Rogers, J. A. Compliant and Stretchable Thermoelectric Coils for Energy Harvesting in Miniature Flexible Devices. *Sci. Adv.* 2018, 4, eaau5849.
31. Jeong, S. H.; Cruz, F. J.; Chen, S.; Gravier, L.; Liu, J.; Wu, Z.; Hjort, K.; Zhang, S. L.; Zhang, Z. B. Stretchable Thermoelectric Generators Metalized with Liquid Silicon. *ACS Appl. Mater. Interfaces 2017*, 9, 15791–15797.
32. Rojas, J. P.; Singh, D.; Conchouso, D.; Arevalo, A.; Foulds, I. G.; Hussain, M. M. Stretchable Helical Architecture Inorganic-Organic Hetero Thermoelectric Generator. *Nano Energy 2016*, 30, 691–699.
33. Wang, Z.; Li, H.; Tang, Z.; Liu, Z.; Ruan, M.; Ma, L.; Yang, Q.; Wang, D.; Zhi, C. Hydrogel Electrolytes for Flexible Aqueous Energy Storage Devices. *Adv. Funct. Mater.* 2018, 28, 1804560.
34. Zhang, L. M.; He, Y.; Cheng, S.; Sheng, H.; Dai, K.; Zheng, W. J.; Wang, M. X.; Chen, Z. S.; Chen, Y. M.; Suo, Z. Self-Healing, Adhesive, and Highly Stretchable Ionogel as a Strain Sensor for Extremely Large Deformation. *Small* 2019, 15, 1804651.

35. Liang, G.; Liu, Z.; Mo, F.; Tang, Z.; Li, H.; Wang, Z.; Sarangi, V.; Pramanick, A.; Fan, J.; Zhi, C. Self-Healable Electroluminescent Devices. *Light Sci Appl* 2018, 7, 102.

36. Zhang, R.; Fu, Q.; Zhou, K.; Yao, Y.; Zhu, X. Ultra Stretchable, Tough and Self-Healable Poly(Acrylic Acid) Hydrogels Cross-Linked by Self-Enhanced High-Density Hydrogen Bonds. *Polymer* 2020, 199, 122603.

37. Liew, C.-W.; Ng, H. M.; Numan, A.; Ramesh, S. Poly(Acrylic Acid)-Based Hybrid Inorganic-Organic Electrolytes Membrane for Electrical Double Layer Capacitors Application. *Polymers* 2016, 8, 179.

38. Jiao, Y.; Lu, K.; Lu, Y.; Yue, Y.; Xu, X.; Xiao, H.; Li, J.; Han, J. Highly Viscoelastic, Stretchable, Conductive, and Self-Healing Strain Sensors Based on Cellulose Nanofiber-Reinforced Polyacrylic Acid Hydrogel. *Cellulose* 2021, 28, 4295–4311.

39. Hu, R.; Ji, G.; Wang, Y.; Zhao, J.; Zheng, J. Rational Design of Multiple Hydrogen Bonds to Improve the Mechanical Property of Rigid Pani. *Extreme Mech. Lett.* 2021, 42, 101136.

40. Kang, D.; Park, N.; Ko, J.-h.; Bae, E.; Park, W. Oxygen-Induced P-Type Doping of a Long Individual Single-Walled Carbon Nanotube. *Nanotechnology* 2005, 16, 1048.

41. Zhou, W.; Fan, Q.; Zhang, Q.; Cai, L.; Li, K.; Gu, X.; Yang, F.; Zhang, N.; Wang, Y.; Liu, H. High-Performance and Compact-Designed Flexible Thermoelectric Modules Enabled by a Reticulate Carbon Nanotube Architecture. *Nat. Commun.* 2017, 8, 1–9.

42. Shim, M.; Javey, A.; Shi Kam, N. W.; Dai, H. Polymer Functionalization for Air-Stable N-Type Carbon Nanotube Field-Effect Transistors. *J. Am. Chem. Soc.* 2001, 123, 11512–11513.

43. Yu, C.; Murali, A.; Choi, K.; Ryu, Y. Air-Stable Fabric Thermoelectric Modules Made of N- and P-Type Carbon Nanotubes. *Energy Environ. Sci.* 2012, 5, 9481–9486.

44. Nunna, R.; Qiu, P.; Yin, M.; Chen, H.; Hanus, R.; Song, Q.; Zhang, T.; Chou, M.-Y.; Agne, M. T.; He, J.; Snyder, G. J.; Shi, X.; Chen, L. Ultrahigh Thermoelectric Performance in Cu2Se-Based Hybrid Materials with Highly Dispersed Molecular CNTs. *Energy Environ. Sci.* 2017, 10, 1928–1935.