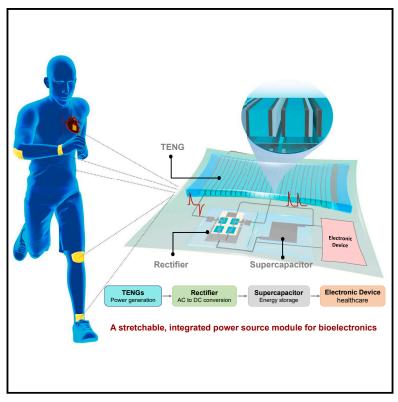
Device

An intrinsically stretchable power-source system for bioelectronics

Graphical abstract



Highlights

- A general design flow for intrinsically stretchable TENGs has been proposed
- Stretchable TENGs achieve superior tissue conformability and high-output power
- Stretchable power regulation circuits have been developed based on transistors
- A stretchable, integrated power-source module for bioelectronics has been developed

Authors

Ping Cheng, Shilei Dai, Youdi Liu, ..., Zhen Wen, Xuhui Sun, Sihong Wang

Correspondence

sihongwang@uchicago.edu

In brief

This work demonstrates a fully stretchable and integrated power source, consisting of a triboelectric nanogenerator (TENG), a polymeric fourtransistor rectifier, and a supercapacitor, designed to harvest stretching-type mechanical energy from tissue/organ motions. The TENG, featuring tissue-like softness, high stretchability, and high output power, has been successfully tested as an implantable harvester on a porcine heart. The fully integrated power source can operate conformably on the human body, offering sustainable energy for wearable and implantable electronics.



Develop Prototype with demonstrated applications in relevant environment Cheng et al., 2024, Device 2, 100216 January 19, 2024 © 2023 Elsevier Inc. https://doi.org/10.1016/j.device.2023.100216



Device



Article

An intrinsically stretchable power-source system for bioelectronics

Ping Cheng,^{1,2,7} Shilei Dai,^{1,7} Youdi Liu,^{1,7} Yang Li,^{1,7} Hidenori Hayashi,³ Rithvik Papani,¹ Qi Su,¹ Nan Li,¹ Yahao Dai,¹ Wei Liu,¹ Huawei Hu,¹ Zixiao Liu,^{4,5} Lihua Jin,⁵ Narutoshi Hibino,³ Zhen Wen,² Xuhui Sun,² and Sihong Wang^{1,6,8,*} ¹Pritzker School of Molecular Engineering, The University of Chicago, Chicago, IL 60637, USA

²Institute of Functional Nano and Soft Materials (FUNSOM), Jiangsu Key Laboratory for Carbon-Based Functional Materials and Devices, Soochow University, Suzhou 215123, China

³Division of Cardiac Surgery, Department of Surgery, University of Chicago, Chicago, IL 60637, USA

⁴Department of Materials Science and Engineering, University of California, Los Angeles, Los Angeles, CA 90095, USA

⁵Department of Mechanical and Aerospace Engineering, University of California, Los Angeles, Los Angeles, CA 90095, USA

⁶Nanoscience and Technology Division and Center for Molecular Engineering, Argonne National Laboratory, Lemont, IL 60439, USA ⁷These authors contributed equally

⁸Lead contact

*Correspondence: sihongwang@uchicago.edu https://doi.org/10.1016/j.device.2023.100216

THE BIGGER PICTURE Integrating bioelectronics with the human body for healthcare necessitates stretchable power sources that are sustainable in energy supply, maintenance-free, and biocompatible. Despite triboelectric nanogenerators (TENGs) having been demonstrated as promising platforms for scavenging energy from tissue/organ motions to power bioelectronics, state-of-the-art stretchable TENGs suffer from limited output performance, lacking stretchable power-management circuits, and rarely achieve energy harvesting from stretching-type deformations. Here, we present a stretchable power module system that harvests energy from body movements. It includes a TENG, a power-rectifying circuit, and a supercapacitor, all designed for over 100% strain stretchability and efficient stretching-type mechanical energy harvesting. This work represents a significant advance in sustainable, biocompatible, and maintenance-free power sources, potentially transforming power supply in wearable and implantable bioelectronics.

SUMMARY

Soft, stretchable electronics, feasible for wear and implantation in the human body, face the challenge of power supply. A promising approach is scavenging energy from body motions, necessitating mechanical-energy-harvesting devices that can stretch with the human body. However, existing stretchable designs have much higher stiffness than biotissues and lack stretchable circuits for power regulation. Here, we report a fully stretchable, integrated power source, consisting of a triboelectric nanogenerator (TENG), a polymeric fourtransistor-based rectifier, and a supercapacitor, that harvests energy from body motions. Guided by rational design and built from ultrasoft elastomers, the TENG achieves tissue-like modulus, high stretchability, and high-output power, which we demonstrated in an implantable harvester on a porcine heart. We also demonstrate that the fully integrated power source, as the first of its kind, can operate conformably on the human body and serve as a sustainable power source for wearable and implantable electronic systems.

INTRODUCTION

The integration of electronics with the human body and other biological systems has recently received extensive developments for personal healthcare, precision medicine, biological studies, medical therapy, and prosthetic e-skins.^{1–4} The overarching goal for the development of such human-integrated electronics is to achieve stable and sustainable long-term operation on or inside the human body, which conjunctionally relies

on two criteria. First, the entire electronic system, including the power sources, must have similar mechanical (i.e., soft and stretchable) properties to skin and biological tissues so as to improve comfort for the wearer, suppress invasive reactions, and enhance signal quality. In this aspect, substantial progress has been made recently in the development of stretchable materials,^{5,6} sensors,^{7–11} transistors,^{12,13} circuits,¹⁴ and displays.^{15–17} Second, the power sources of the electronics, which have been conventionally served by rigid batteries, should be

CelPress



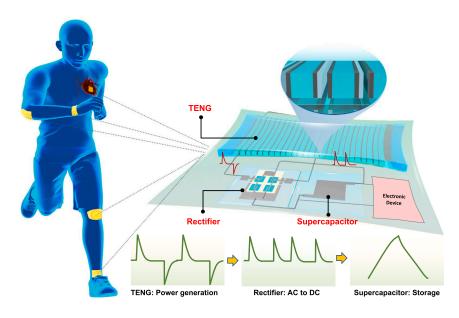


Figure 1. Schematic of a fully stretchable and fully integrated power module

The system consists of a stretchable TENG, a stretchable rectifying circuit, and a stretchable supercapacitor, which can serve as a sustainable and human-compatible power source for wearable and implantable electronics.

tronics will be the trend of wearable/ implantable power sources.

In the human body, mechanical energy is the most ubiquitous energy type, which mostly exists in the form of muscle and tissue stretching from body movements or organ deformations. In recent years, triboelectric nanogenerators (TENGs) have been developed as a highly potent technology for harvesting irregular and low-frequency mechanical energy,^{27–32} with the advantages of high efficiency, broad material choices, and lightweight construction.

sustainable in energy supply and maintenance free, ideally with an unlimited lifetime.^{18–20} This is especially important for implantable devices (such as pacemakers and neurostimulators) for alleviating additional surgical procedures for battery replacement. However, much less progress has been made in this second aspect.

The paths to realizing such sustainable power supplies mainly include two general options: wireless power delivery²¹⁻²³ and bioenergy harvesting.^{19,24} Since the wireless approach can cause tissue damage and is only limited to the near-epidermis regions, bioenergy harvesting is thought to be more generally applicable. For this, skin- and tissue-like stretchability is one essential requirement for achieving adequate biocompatibility and needs to be combined not only with high-efficiency energy-harvesting devices but also with power management components that convert fluctuating, sometimes alternating-current (AC) and pulsed power, into a constant-voltage supply for satisfying the needs of electronics.^{25,26}

In this regard, self-powered implantable devices that scavenge energy from the human body are attractive for long-term utility. To date, stretchable electronics are almost always powered by rigid batteries, which greatly reduces their compatibility with our skin/tissues. Thus, it is critical to develop a new power source that incorporates skin-like properties, such as stretchability and long-term durability. Some presented stretchable batteries, like lithium-ion batteries, still require replacing the drained battery after a period of time. However, it is invasive to remove the implantable batteries by surgery. Therefore, the novel power source still needs to have the ability to generate the energy itself, obviating the need for secondary device removal for medical implants. Coincidentally, the stretching movement of stretchable electronics on our skin/tissue is exactly a potential mechanical energy that can be collected to power wearable electronics. Designing a self-powered stretchable power source for harvesting existing stretching energy to supply stretchable elec-

retching energy to supply stretchable elec- th

For the use of TENGs for harvesting human body energy,³³⁻³ the impartment of stretchability is particularly beneficial for two main reasons. First, it will significantly enhance skin/tissue conformability and biocompatibility. Second, as tissue/muscle stretching deformation is arguably the most common type of mechanical energy in the human body; the larger stretchability of TENGs can ensure the full harnessing of stretching deformation as the energy input for power generation. Although there have been some reports of stretchable designs for TENGs,^{36–41} the utilized materials and device structures significantly limit the output performance and rarely achieve energy harvesting from stretching-type deformations, which together manifest a significant gap toward practical utilization for powering human-integrated electronics. In addition, there have not vet been successful examples of imparting stretchability to the indispensable power management circuits, which should at least include a rectifier and a capacitor or battery, which is critical to the development of a fully stretchable TENG-based power module.

Herein, we present the first intrinsically stretchable power module system based on mechanical energy harvesting from the human body (Figure 1), which includes a high-output stretchable TENG, a stretchable power-rectifying circuit, and a stretchable supercapacitor. For the stretchable TENG, we created a rational design that not only achieves stretchability beyond 100% strain and tissue-like softness but also realizes highly efficient transmission and conversion of the stretching energy from the tissue/skin surface. Compared to all reported energy harvesting from stretching, our TENG realized a significant improvement in output power density by two orders of magnitude (Table S1). Most importantly, our stretchable TENG has been attached to a live porcine heart and converts the expansioncontraction of the cardiac muscle during heart beating into electricity. Our intrinsically stretchable rectifier built by four stretchable thin-film transistors (TFTs) can effectively convert the pulsed AC output generated by the TENG into direct-current

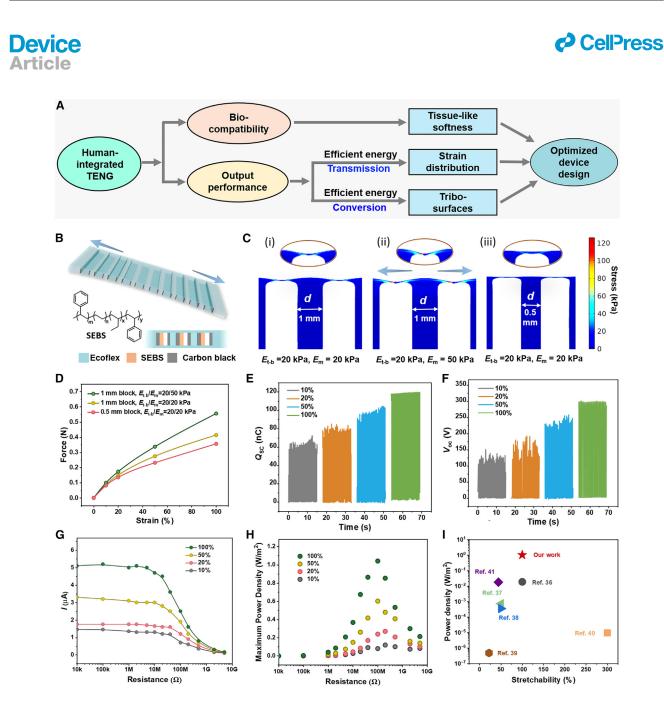


Figure 2. Design and characterization of the stretchable TENG using finite-element analysis

(A) Our proposed design flow for stretchable human-integrated TENGs.

(B) Schematic of the intrinsically stretchable TENG and the chemical structure of SEBS.

(C) Finite-element analysis (FEA) results of the stress distributions in three model TENGs under 100% strain, which shows the influence of the moduli of the middle layer and the block thickness.

(D) Extracted force-strain curves of the three simulated TENG structures.

(E–H) Transferred charges (E), open-circuit voltage (F), resistance-dependent current (G), and peak power density (H) from the TENG with the optimized design. (I) Comparison of stretching-generated peak power density and stretchability of our TENG with the other reported stretchable TENGs, showing that our TENG can simultaneously achieve high output density and high stretchability.

(DC) output without any observable loss of energy. Then, the rectified power is stored in a stretchable supercapacitor that is also integrated with the power module. We believe that the successful impartment of stretchability to all the major components of an energy-harvesting-based power module is a landmark result in the development of sustainable and biocompatible power sources for bioelectronics.

RESULTS

For the design of stretchable TENGs for biointegrations, there are two major considerations (Figure 2A): biocompatibility and output performance. Mechanically, the biocompatibility of an implanted device requires a tissue-like modulus in the range of 1–50 kPa.⁴² To achieve such an ultra-low modulus, we

CellPress

Device Article

devised a meta-structure-type device design with a series of embedded gaps (Figure 2B) for easier deformation under stretching and built it with a type of ultra-soft elastomer, Ecoflex. We chose Ecoflex because of its low modulus, good biocompatibility, and commercial availability. Each gap in the meta-structure houses a contact-separation-mode TENG structure with a pair of electrodes and triboelectric surfaces (Figures S1 and S2).

As the other branch of the design flow (Figure 2A), the output performance of the stretchable TENG (Figure 2B) when harvesting stretching energy is jointly determined by the two steps of the mechanical-energy-harvesting process: the transmission of mechanical energy from an external resource (e.g., skin or organ surface) to the opening of the gaps and then the energy conversion to electricity by the embedded TENG structure. The efficiency of the first step can be maximized by ensuring that the surface connection layers have a much smaller stretching stiffness than the middle structural blocks so that the applied strain mostly converts to the separation of the triboelectric layers in the unit cells. The efficiency of the second step-electricity generation from the TENG unit cells-is mainly determined by the two triboelectric surfaces, which need to have high triboelectric charge density and low adhesion. In our design, these two requirements are concurrently met by a type of elastomer, styrene-ethylene-butadiene-styrene (SEBS; Figures S3 and S4), when the conductive composite of Ecoflex and carbon black is used as the counter layer.

Structurally, the main design parameters influencing the stretching energy distribution on the TENG are the modulus ratio between the top/bottom connecting layers and the middle block (with their moduli labeled as E_{t-b} and E_m , respectively) as well as the thickness (d) of the middle blocks. We performed finiteelement analysis (FEA) on different TENG models to help us better understand the quantitative influences of these parameters (Figures 2C, S5, and S6). With the same global strain of 100%, the stress distributions on different "TENGs" are plotted in Figure S7. Obviously, higher E_m leads to increased local stress at the opening tips of the top/bottom layers, which results in a higher stretching force for deforming the TENG (Figures 2C, S5, and S7A). This will lower the energy conversion efficiency. Therefore, a lower modulus for the middle blocks is favorable. On the other hand, when the block thickness d decreases while other geometric and mechanical parameters are maintained, the strain-concentration effect on the top/bottom layers decreases, thereby resulting in the slightly decreased local stress at the opening tips of the top/bottom layers (Figures 2C, S6, and S7B). As such, a relatively smaller d can lower the stretching force (Figure 2D) but has almost no effect on the transferred charges and open-circuit voltage of the TENG (Figure S8).

Under this guidance, we created the design experimentally using Ecoflex for the entire structure (i.e., 20 kPa for both E_{t-b} and E_m), with the dimensions of 30 mm length with 10 TENG subunits, 5 mm thickness, and 8 mm width. Under 100% strain, the mechanical load created by our device is as low as 0.35 N (Figure 2D). When repeated stretching up to 100% strain along the longitudinal direction is applied to the TENG, both the short-circuit charge transfer (Q_{SC} ; Figure 2E) and open-circuit voltage (V_{CC} ; Figure 2F) increase monotonically with the increase of the strain. This trend is also reflected in the current output (Figure 2G) and the areal power density (Figure 2H) generated on external loads with different resistances. In particular, under 100% strain, the Q_{SC} and V_{OC} are about 120 nC and 300 V, respectively, which give rise to a maximum power density of 1.1 W/m² on an external load of 100 M Ω . Compared to all previously reported stretchable TENGs for harvesting stretching energy,^{36–41} our device not only improves the power density by at least two orders of magnitude but also has the highest stretchability and softness (Figure 2I; Table S1). As such, our design greatly advances the applicability of skin-/tissue-like TENGs for powering wearable and implantable electronics. Our TENG also showed good long-term operational stability, as shown by repeated stretching between 0% and 100% strain more than 10,000 times (Figure S9). As the stretching cycle increases, the output performance of our stretchable TENGs had some slight increase (~20%), which is due to the amount of charge generated by friction still gradually accumulating. The effect of stretching speed on the performance of TENGs was also investigated (Figure S10), and it was found that the influence is mainly on the current output. It should also be noted that this same design concept of a "soft meta-structure" for achieving efficient harvesting of stretching energy can also be utilized in device designs that can provide biaxial stretching energy harvesting (Figure S11).

To test our stretchable TENG's capacity for generating power from the human body, we attached the device to different parts on and inside the human/animal body. First, for daily body motions such as walking, jogging, and hand movements, which mainly happen through joint movements, we have attached our stretchable TENGs to the wrist, knee, and ankle areas (Figures 3A-3C and S12). The skin-like softness and stretchability enable highly effective harvesting of the mechanical energy that exists in the form of joint flexing and skin stretching, which generates an output performance of nearly 180 V in V_{OC} and 2.0 μ A in short-circuit current (I_{SC})-enough to power 50 LEDs (Videos S1, S2, and S3). The capability of our stretchable TENG to serve as the implantable power source through harvesting energy from organ motions is tested by suturing the TENG to a living adult porcine heart (Figures 3D and 3E). As shown in Video S4, benefiting from the tissue-level softness of the TENG, the cyclic strain of ca. 20% on the cardiac muscle during the heartbeats can be transformed effectively onto the stretchable TENG that is sutured onto the left ventricle. The generated power outputs are shown in Figures 3F-3H. Compared to the reported cardiac-energy-harvesting devices built with nonstretchable and high-modulus materials (such as polyimide) (Table S2), the unprecedented tissue-level modulus (~20 kPa) that is realized in our TENG can minimize the mechanical load (i.e., only 0.35 N is required to achieve 100% strain; Figure 2D) and areal constraint added onto the beating heart, which is highly preferred for the long-term wellbeing of the heart.

The utilization of the output from the TENG for powering electronics requires the conversion of the AC to DC output by a rectifier circuit so that it can be stored in a capacitor or battery to provide stable output. So far, the most typical rectifier design that has been utilized for TENGs is the bridge rectifier built with four diodes, which can provide full-wave rectification to AC

Device Article

CellPress

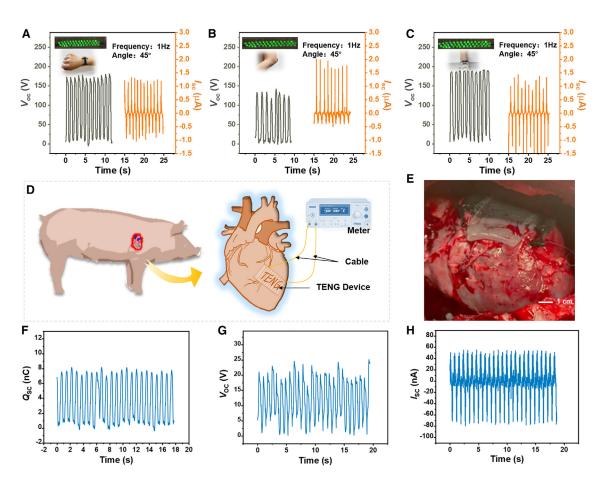


Figure 3. Proof-of-concept demonstrations of the stretchable TENG for harvesting biomechanical energy from the human body and the porcine heart

(A-C) Open-circuit voltage and short-circuit current curves of the TENG for harvesting the energy on the (A) wrist, (B) knee, and (C) ankle.

(D) Schematic of a stretchable TENG implanted into a pig to harvest energy from cardiac muscle stretching during heart beating.

(E) Photograph showing the attachment of a TENG to a live porcine heart.

(F–H) In vivo measurement results of the transferred charge (F), open-circuit voltage (G), and short-circuit current (H) generated by the heart beating.

inputs. Actually, diode-like performance can be obtained from a TFT by connecting the drain and gate electrodes. In comparison with the recently reported stretchable vertical-stacked-diodebased rectifiers,⁴³ the use of stretchable TFTs to implement rectifiers not only achieves higher stretchability but also gives higher operation voltage. Both of these are highly important attributes for the power regulation of stretchable TENGs. Here, using a previously reported stretchable TFT design,⁴⁴ we successfully created the first stretchable full-wave rectifier by connecting four stretchable TFTs (Figure 4A) with the design shown in Figure 4D. The stretchable TFT is built with the CONPHINE design⁴⁴ of PDPP-TT ([poly-[2,5-bis(2-octyldodecyl)-3,6-di(thiophen-2-yl)pyrrolo[3,4-c]pyrrole-1,4(2H,5H)-dionel-alt-thieno[3,2b]thiophene]) as the channel layer, SEBS (1.8 µm thick) as the dielectric layer, and carbon nanotube (CNT) assemblies as the source (S), drain (D), and gate (G) electrodes. In the nonstretched state, the standard transfer curve of the transistor shows an ideal switching behavior with an on/off ratio of 10⁵ (Figure 4B). When the D and G electrodes are connected, the operation of the transistor as a two-terminal device indeed gives diode-like rectifying behavior to the input voltage, 45,46 with an on/off ratio also close to 10⁵ (Figure 4C). During the stretching to 100% strain and upon release, both the transfer curves of the transistor and the current-voltage (I-V) curves of the transistor-based diode maintain high stability, with the decrease of the on-current well within one order of magnitude (Figures 4B and 4C). The performance change of the stretchable TFTbased diode under strain mainly stems from the synergistic effects of mobility change and device size change during the stretching process (Figures S13 and S14; Notes S1 and S2). When four such transistors are fabricated on the same stretchable substrate (Figure 4D) with the circuit connection shown in Figure 4E, a stretchable bridge rectifier (Figure 4G) is created to rectify the power output from our stretchable TENG. As shown in Figures 4F, S15, and S16, the initial AC output gets rectified in full wave to the DC output without any observable loss of amplitude before or after mechanical deformation (e.g., stretching and twisting). This shows the sufficient capability of this stretchable rectifier to serve as an ideal power regulator for the stretchable TENG. For the use of such stretchable

CellPress

Device Article

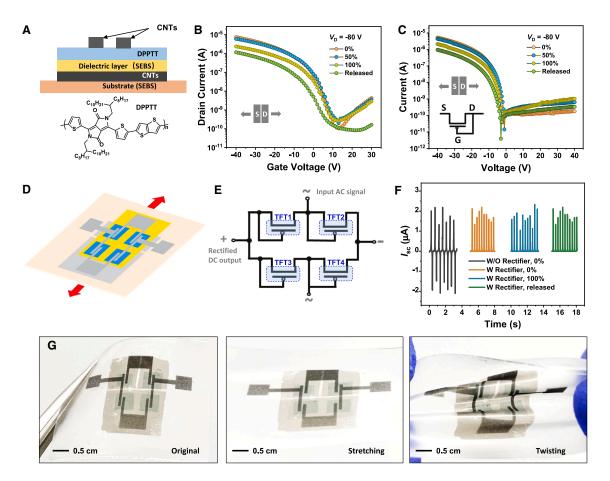


Figure 4. Intrinsically stretchable bridge rectifier made from stretchable thin-film transistors (TFTs)

(A) The device structure of the stretchable TFTs, its stretchable components, and the chemical structure of DPP-TT.

(B) Transfer curves measured from the transistor during stretching to 100% strain.

(C) The stretchable transistor operates as a stretchable diode by connecting the gate and drain electrodes together, with the rectifying current-voltage (*I-V*) curves during the stretching from 0% to 100% strain.

(D and E) Schematic illustration of the stretchable bridge rectifier made from four stretchable transistors for the full-wave rectification of AC output to DC output (D) and the circuit diagram (E).

(F) Short-circuit currents (*I*_{SC}) from a stretchable triboelectric nanogenerator, before and after the rectification by the stretchable bridge rectifier under different strains.

(G) Photographs of the stretchable bridge rectifier under various deformations.

rectifiers on TENGs with lower voltage output, the turn-on voltage of the rectifier can be further lowered by fabricating stretchable dielectric layers using materials with higher dielectric constant⁴⁷ or film deposition methods⁴⁸ that give denser morphology to realize lower thickness.

Finally, a fully stretchable power source system based on mechanical energy harvesting is demonstrated by integrating a stretchable TENG, a stretchable bridge rectifier, and a stretchable supercapacitor (Figure 5). In particular, the stretchable supercapacitor is built with CNT electrodes, a phosphoric acid/PVA (polyvinyl alcohol) gel electrolyte, and SEBS encapsulation. Its energy storage performance is shown by cyclic voltammetry (CV) with different scan rates (Figure S17A) and galvanostatic charging-discharging curves at different currents (Figure S17B). Our supercapacitor can tolerate more than 100 cycles of charging-discharging at 25 μ A without any noticeable

performance degradation (Figure S17C). Its high stretchability ensures unaffected performance during the stretching cycle to 100% strain (Figures 5B and 5C). The stable CV and charging/ discharging curves during stretching may be attributed to (1) the unchanged loading amount of CNTs during stretching, (2) the almost unchanged electrolyte impedance, and (3) the decent adhesion between the electrolyte and the CNTs (Figures S18 and S19). To further demonstrate the function of a full power source module that harvests human-motion energy, the stretchable TENG, bridge rectifier, and supercapacitor are integrated into the same piece of PDMS substrate, with the connections made by a stretchable silver paste (Figure 5E). When the entire power system is conformably attached to the human wrist, the bending motions of the wrist can be converted to electrical energy through the stretchable TENG part without affecting the function of the bridge rectifier and the

Device Article

CellPress

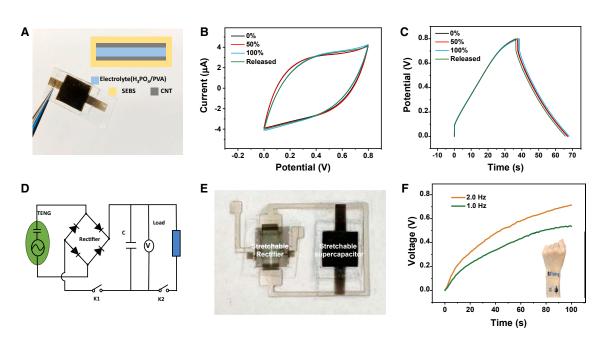


Figure 5. Intrinsically stretchable power source module system for harvesting stretching energy from the human body (A) Device structure and picture of the stretchable supercapacitor.

(B and C) CV (B) and galvanostatic charge/discharge (GCD) (C) curves of the stretchable supercapacitor under stretching.

(D and E) Connection circuit (D) and picture of the integrated power source module (E).

(F) Energy stored in the power source module worn on the human wrist harvesting energy from the wrist's bending motion.

supercapacitor. As such, the mechanical energy from the human body gets efficiently harnessed, rectified, and ultimately stored in the supercapacitor (Figure 5F), which can be used for powering electronic systems. Future research can further parallelly connect a stretchable diode⁴⁹ with supercapacitors in the circuit to ensure that the voltage applied to the supercapacitors does not exceed operating limits, which may require optimizing the diode's turn-on voltage.

DISCUSSION

A mechanical-energy-harvesting-based power source module needs to meet three requirements to utilize body motions to sustainably power human-integrated electronics: (1) skin-/tissuelike mechanical compatibility, (2) high efficiency in harnessing body-motion-type energy, and (3) integrated power regulation for providing DC output. For the first two aspects, we proposed a general design flow for intrinsically stretchable TENGs for harvesting the most prevalent type of mechanical deformationsstretching-from the human body. The device structure we created achieves both superior skin/tissue conformability and unprecedentedly high output power for harvesting stretching energy up to 100% strain. Its practicability as a body-motion-based power source is demonstrated through both on-skin operations on joints and implanted operations on a live porcine heart. Moreover, for the third aspect of integrated power regulation, we established the first stretchable design by taking advantage of the state-of-the-art advancement in stretchable polymer transistors, which can provide full-wave rectification to the AC output from TENGs. This further enabled the realization of a fully integrated, fully stretchable power source module that can constantly produce DC output by harnessing body motions. Along the path laid out by this work, we expect that the power output can be further improved by optimizing the triboelectric materials for achieving higher triboelectric charge density and refining the fabrication for enhancing the utilization of the mechanical energy input. This work presents an important step in solving the powersupply challenge for realizing the sustainable operation of wearable/implantable electronics.

EXPERIMENTAL PROCEDURES

Resource availability

Lead contact

Further information and requests for resources should be directed to and will be fulfilled by the lead contact, Sihong Wang (sihongwang@uchicago.edu). *Materials availability*

All processing solvents, such as chlorobenzene, toluene, absolute ethanol, and 2-propanol, were purchased from commercial sources (Sigma-Aldrich) and used as received. Super P Conductive Carbon Black was purchased from MTI. Ecoflex-010, -030, and -050 were purchased from Smooth-On. The polymer semiconductor PDPP-TT was synthesized according to the reported method.⁴⁴ The SEBS compounds H1221 and H1052, with poly(ethylene-co-butylene) volume fractions of, respectively, 88% and 80%, were provided by Asahi Kasei. SEBS rubbers have been reported to have good long-term stability and biocompatibility.⁵⁰ We used SEBS H1052 as a stretch-able dielectric layer and SEBS H1221 in the CONPHINE semiconductor. Dextran was purchased from Sigma-Aldrich and used as received. CNTs for electrodes were purchased from Carbon Solutions (P3-SWCNTs [single-walled CNTs]). The silver paste 126-49 was purchased from Creative Material. Other commercial reactants were purchased from Sigma-Aldrich and used without further purification.

CellPress

Data and code availability

The data that support the findings of this study are available within the paper and its supplemental information files. Additional data and files are available from the corresponding author upon reasonable request.

Fabrication of the stretchable TENG

First, a 200-µm-thick PET film was coated with Dextran on both sides (1,000 rpm for 30 s). Next, SEBS solution (100 mg/mL in toluene) was spin coated on one surface of the Dextran-PET at a speed of 1,000 rpm for 30 s and then baked in an oven at 80°C for 5 min. Next, the mixture of Ecoflex-030 and carbon black (10 wt %) was coated on both sides of this film as electrodes. Then, the film was cut into ten electrodes with a dimension of 4 \times 30 mm (width × length). The ten units were put on a mold, and 0.5 g Ecoflex-010 was poured on top as the bottom layer. After baking at 80°C for 5 min in the oven, another 4.5 g Ecoflex-050 was poured into the mold and baked for another 8 min. Then, the last 0.5 g Ecoflex-010 was poured into the mold as the top layer and covered by an acrylic plate. After baking for 30 min, the device was soaked in water for 3 h to remove the PET films, which gave the TENG devices. For the in vivo test, the TENGs were encapsulated by using Ecoflex to make sure that the device could function properly in the bleeding pig heart. To avoid the creation of negative pressure inside the encapsulation and maintain uniform pressure distribution, we left a small hole on each block between adjacent TENG array units and added a thin cavity on one sidewall of the entire TENG device.

Fabrication of the stretchable rectifiers

The G electrode was fabricated on a 4 inch Si/SiO₂ wafer that was sonicated in acetone, 2-propanol, and deionized water sequentially for 5 min, each followed by blow drying with nitrogen gas. The wafer was then treated with oxygen plasma (350 W, 200 mTorr) for 3 min. The CNTs (P2-SWCNTs) serving as the G electrode were first dispersed in N-methylpyrrolidone (NMP; 20 mg in 70 mL), sonicated with a probe sonicator for 30 min, and centrifuged at 8,000 rpm for 20 min. The supernatant was collected for spray coating on the wafer at 180°C on a hot plate. The SEBS 1062 in toluene (10 mg/mL) was spin coated on the P2-SWCNT-coated wafer at 1,000 rpm for 1 min. The SEBS substrate was prepared by dispersing SEBS 1062 in toluene (200 mg/mL) that was drop casted on a glass substrate covered by glassware for slow evaporation of the solvent in 2 days. The SEBS substrate was used to peel off the P2-SWCNTs from the Si/SiO₂ wafer.

To make the dielectric layer, SEBS 1052 was dispersed in toluene (60 mg/mL) and spin coated on octadecyltrimethoxysilane (OTS)-treated Si wafer at 1,000 rpm for 1 min. The superhydrophobic Si wafer allows the ease of transfer of the SEBS 1052 dielectric layer to the CNT G electrode by a PDMS stamp. The stretchable semiconductor films were made by blending PDPP-TT with SEBS 1221 in chlorobenzene, with a weight ratio of 1:1 and a total concentration of 10 mg/mL. The solution was spin coated on the OTS-treated Si wafer at 1,000 rpm for 1 min, followed by annealing at 150°C for 30 min. The semiconducting film was then transferred by a PDMS stamp to the dielectric layer.

Top S/D electrodes were patterned by spray coating P3-SWCNT solution through a shadow mask (Invar). The CNT solution was prepared by dispersing 20 mg P3-SWCNT in 70 mL 2-propanol with two drops of water through a process of consecutive 3 h bath sonications, 10 min tip sonication, and then centrifugation at 6,000 rpm for 20 min. The P3-SWCNT dispersion was spray coated on the semiconductor at 75°C to create patterns of S and D electrodes.

Fabrication of the stretchable supercapacitor

First, PVA/H₃PO₄ gel electrolyte was prepared as follows: 5 g H₃PO₄ and 5 g PVA powder were sequentially added into 50 mL deionized water. The mixture was heated to 85°C under stirring until the solution became clear. CNTs were sprayed on two SEBS 1052 substrates as electrodes (size: 1 × 1 cm). A SEBS 1221 spacer was sandwiched between two SEBS-supported electrodes, with the PVA/H₃PO₄ gel electrolyte added into the middle gap. Then, the gaps between the spacer and the top and bottom SEBS substrates were sealed by injecting SEBS solution. Then, after the complete solidification of the PVA/H₃PO₄ gel, the stretchable supercapacitor was ready for testing.



Assembly of the power source module system

A piece of 1-mm-thick PDMS film was used as the common substrate, on which stretchable silver paste 126-49 was blade coated and patterned through a shadow mask as the interconnects. Next, the PDMS was plasma treated, and the fabricated rectifier and supercapacitor were then physically attached to the PDMS substrate by pressing. To prevent delamination, another piece of 1-mm-thick PDMS was used to cover the circuit as the encapsulation. To further enhance the adhesion force between each layer, the uncured PDMS solution can be coated on the surface of each layer before assembly and cured at room temperature after assembly. The stretchable TENG was connected to the rectifier through two copper wires after fabrication.

SUPPLEMENTAL INFORMATION

Supplemental information can be found online at https://doi.org/10.1016/j. device.2023.100216.

ACKNOWLEDGMENTS

This work is supported by the start-up fund from the University of Chicago and the China Scholarship Council (CSC). The authors thank the University of Chicago Animal Resources Center (RRID: SCR_021806), especially the Carlson Large Animal Clinic Staff, Dr. Allison Ostdiek, Dr. Darya Mailhiot, Jenny McGrath, Alyssa Brown, Erika Becerra, and Pierre Latalladi for their assistance with animal surgery and housing.

AUTHOR CONTRIBUTIONS

S.W. conceived the original idea and supervised this study. P.C. and S.W. designed the experiments. P.C., Y. Liu, S.D., and Y. Li. performed the material characterizations, device fabrications, and measurements. P.C. and Z.L. performed the simulation. P.C. and Y.Liu fabricated the stretchable rectifier. P.C. and S.D. set up the stretchable rectifier testing system and performed the rectifier device measurement. N.L. helped with the organic semiconductor synthesis, and Q.S. helped with the demo shooting. Y.L., Y.D., W.L., and H.H. helped with the data analyses. P.C., S.W., S.D., and Y.Liu wrote the manuscript. All authors reviewed and commented on the manuscript.

DECLARATION OF INTERESTS

A patent application has been filed by the University of Chicago.

Received: November 22, 2022 Revised: November 17, 2023 Accepted: November 21, 2023 Published: December 19, 2023

REFERENCES

- Wang, C., Qi, B., Lin, M., Zhang, Z., Makihata, M., Liu, B., Zhou, S., Huang, Y.-h., Hu, H., Gu, Y., et al. (2021). Continuous monitoring of deep-tissue haemodynamics with stretchable ultrasonic phased arrays. Nat. Biomed. Eng. 5, 749–758.
- Jung, Y.H., Yoo, J.-Y., Vázquez-Guardado, A., Kim, J.-H., Kim, J.-T., Luan, H., Park, M., Lim, J., Shin, H.-S., Su, C.-J., et al. (2022). A wireless haptic interface for programmable patterns of touch across large areas of the skin. Nat. Electron. 5, 374–385.
- Ates, H.C., Yetisen, A.K., Güder, F., and Dincer, C. (2021). Wearable devices for the detection of COVID-19. Nat. Electron. 4, 13–14.
- Kim, D.-H., and Kim, D.C. (2018). Stretchable electronics on another level. Nat. Electron. 1, 440–441.
- Dai, Y., Dai, S., Li, N., Li, Y., Moser, M., Strzalka, J., Prominski, A., Liu, Y., Zhang, Q., Li, S., et al. (2022). Stretchable Redox-Active Semiconducting Polymers for High-Performance Organic Electrochemical Transistors. Adv. Mater. 34, 2201178.

Device Article



- Lv, J., Thangavel, G., Li, Y., Xiong, J., Gao, D., Ciou, J., Tan, M.W.M., Aziz, I., Chen, S., Chen, J., et al. (2021). Printable elastomeric electrodes with sweat-enhanced conductivity for wearables. Sci. Adv. 7, eabg8433.
- Su, Q., Zou, Q., Li, Y., Chen, Y., Teng, S.-Y., Kelleher, J.T., Nith, R., Cheng, P., Li, N., Liu, W., et al. (2021). A stretchable and strain-unperturbed pressure sensor for motion interference–free tactile monitoring on skins. Sci. Adv. 7, eabi4563.
- Chen, S., Sun, L., Zhou, X., Guo, Y., Song, J., Qian, S., Liu, Z., Guan, Q., Meade Jeffries, E., Liu, W., et al. (2020). Mechanically and biologically skin-like elastomers for bio-integrated electronics. Nat. Commun. *11*, 1107.
- Hu, S., Chang, S., Xiao, G., Lu, J., Gao, J., Zhang, Y., and Tao, Y. (2022). A Stretchable Multimode Triboelectric Nanogenerator for Energy Harvesting and Self-Powered Sensing. Adv. Mater. Technol. 7, 2100870.
- Lee, Y., Cha, S.H., Kim, Y.W., Choi, D., and Sun, J.Y. (2018). Transparent and attachable ionic communicators based on self-cleanable triboelectric nanogenerators. Nat. Commun. 9, 1804.
- 11. Bariya, M., Nyein, H.Y.Y., and Javey, A. (2018). Wearable sweat sensors. Nat. Electron. 1, 160–171.
- Wang, S., Xu, J., Wang, W., Wang, G.-J.N., Rastak, R., Molina-Lopez, F., Chung, J.W., Niu, S., Feig, V.R., Lopez, J., et al. (2018). Skin electronics from scalable fabrication of an intrinsically stretchable transistor array. Nature 555, 83–88.
- Dai, S., Dai, Y., Zhao, Z., Xia, F., Li, Y., Liu, Y., Cheng, P., Strzalka, J., Li, S., Li, N., et al. (2022). Intrinsically stretchable neuromorphic devices for onbody processing of health data with artificial intelligence. Matter 5, 3375–3390.
- Ma, Z., Huang, Q., Xu, Q., Zhuang, Q., Zhao, X., Yang, Y., Qiu, H., Yang, Z., Wang, C., Chai, Y., and Zheng, Z. (2021). Permeable superelastic liquidmetal fibre mat enables biocompatible and monolithic stretchable electronics. Nat. Mater. 20, 859–868.
- Kim, J.-H., and Park, J.-W. (2021). Intrinsically stretchable organic lightemitting diodes. Sci. Adv. 7, eabd9715.
- Lee, Y., Chung, J.W., Lee, G.H., Kang, H., Kim, J.-Y., Bae, C., Yoo, H., Jeong, S., Cho, H., Kang, S.-G., et al. (2021). Standalone real-time health monitoring patch based on a stretchable organic optoelectronic system. Sci. Adv. 7, eabg9180.
- 17. Yin, D., Feng, J., Ma, R., Liu, Y.-F., Zhang, Y.-L., Zhang, X.-L., Bi, Y.-G., Chen, Q.-D., and Sun, H.-B. (2016). Efficient and mechanically robust stretchable organic light-emitting devices by a laser-programmable buckling process. Nat. Commun. 7, 11573.
- Li, Y., Li, N., De Oliveira, N., and Wang, S. (2021). Implantable bioelectronics toward long-term stability and sustainability. Matter 4, 1125–1141.
- Yu, Y., Nassar, J., Xu, C., Min, J., Yang, Y., Dai, A., Doshi, R., Huang, A., Song, Y., Gehlhar, R., et al. (2020). Biofuel-powered soft electronic skin with multiplexed and wireless sensing for human-machine interfaces. Sci. Robot. 5, eaaz7946.
- Ouyang, H., Liu, Z., Li, N., Shi, B., Zou, Y., Xie, F., Ma, Y., Li, Z., Li, H., Zheng, Q., et al. (2019). Symbiotic cardiac pacemaker. Nat. Commun. *10*, 1821–1910.
- Lee, D.-M., Rubab, N., Hyun, I., Kang, W., Kim, Y.-J., Kang, M., Choi, B.O., and Kim, S.-W. (2022). Ultrasound-mediated triboelectric nanogenerator for powering on-demand transient electronics. Sci. Adv. 8, eabl8423.
- Hinchet, R., Yoon, H.-J., Ryu, H., Kim, M.-K., Choi, E.-K., Kim, D.-S., and Kim, S.-W. (2019). Transcutaneous ultrasound energy harvesting using capacitive triboelectric technology. Science 365, 491–494.
- 23. Choi, Y.S., Yin, R.T., Pfenniger, A., Koo, J., Avila, R., Benjamin Lee, K., Chen, S.W., Lee, G., Li, G., Qiao, Y., et al. (2021). Fully implantable and bioresorbable cardiac pacemakers without leads or batteries. Nat. Biotechnol. 39, 1228–1238.

- Hasan, M.N., Sahlan, S., Osman, K., and Mohamed Ali, M.S. (2021). Energy Harvesters for Wearable Electronics and Biomedical Devices. Adv. Mater. Technol. 6, 2000771.
- Cheng, X., Tang, W., Song, Y., Chen, H., Zhang, H., and Wang, Z.L. (2019). Power management and effective energy storage of pulsed output from triboelectric nanogenerator. Nano Energy 61, 517–532.
- Wang, Z., Wu, Y., Jiang, W., Liu, Q., Wang, X., Zhang, J., Zhou, Z., Zheng, H., Wang, Z., and Wang, Z.L. (2021). A Universal Power Management Strategy Based on Novel Sound-Driven Triboelectric Nanogenerator and Its Fully Self-Powered Wireless System Applications. Adv. Funct. Mater. *31*, 2103081.
- Zhang, C., He, L., Zhou, L., Yang, O., Yuan, W., Wei, X., Liu, Y., Lu, L., Wang, J., and Wang, Z.L. (2021). Active resonance triboelectric nanogenerator for harvesting omnidirectional water-wave energy. Joule 5, 1613–1623.
- Wang, Z.L., and Wang, A.C. (2019). On the origin of contact-electrification. Mater. Today 30, 34–51.
- Wu, C., Wang, A.C., Ding, W., Guo, H., and Wang, Z.L. (2018). Triboelectric Nanogenerator: A Foundation of the Energy for the New Era. Adv. Energy Mater. 9, 1802906.
- Wang, Z.L. (2018). Nanogenerators, self-powered systems, blue energy, piezotronics and piezo-phototronics – A recall on the original thoughts for coining these fields. Nano Energy 54, 477–483.
- Wang, S., Xie, Y., Niu, S., Lin, L., and Wang, Z.L. (2014). Freestanding triboelectric-layer-based nanogenerators for harvesting energy from a moving object or human motion in contact and non-contact modes. Adv. Mater. 26, 2818–2824.
- 32. Cheng, P., Guo, H., Wen, Z., Zhang, C., Yin, X., Li, X., Liu, D., Song, W., Sun, X., Wang, J., and Wang, Z.L. (2019). Largely enhanced triboelectric nanogenerator for efficient harvesting of water wave energy by soft contacted structure. Nano Energy 57, 432–439.
- 33. Yang, Y., Sun, N., Wen, Z., Cheng, P., Zheng, H., Shao, H., Xia, Y., Chen, C., Lan, H., Xie, X., et al. (2018). Liquid-Metal-Based Super-Stretchable and Structure-Designable Triboelectric Nanogenerator for Wearable Electronics. ACS Nano 12, 2027–2034.
- Pu, X., Guo, H., Chen, J., Wang, X., Xi, Y., Hu, C., and Wang, Z.L. (2017). Eye motion triggered self-powered mechnosensational communication system using triboelectric nanogenerator. Sci. Adv. 3, e1700694.
- 35. Yin, L., Kim, K.N., Lv, J., Tehrani, F., Lin, M., Lin, Z., Moon, J.-M., Ma, J., Yu, J., Xu, S., and Wang, J. (2021). A self-sustainable wearable multimodular E-textile bioenergy microgrid system. Nat. Commun. 12, 1542.
- 36. Li, J., Kang, L., Long, Y., Wei, H., Yu, Y., Wang, Y., Ferreira, C.A., Yao, G., Zhang, Z., Carlos, C., et al. (2018). Implanted Battery-Free Direct-Current Micro-Power Supply from in Vivo Breath Energy Harvesting. ACS Appl. Mater. Interfaces 10, 42030–42038.
- Zou, Y., Tan, P., Shi, B., Ouyang, H., Jiang, D., Liu, Z., Li, H., Yu, M., Wang, C., Qu, X., et al. (2019). A bionic stretchable nanogenerator for underwater sensing and energy harvesting. Nat. Commun. *10*, 2695.
- 38. Lim, G.-H., Kwak, S.S., Kwon, N., Kim, T., Kim, H., Kim, S.M., Kim, S.-W., and Lim, B. (2017). Fully stretchable and highly durable triboelectric nanogenerators based on gold-nanosheet electrodes for self-powered humanmotion detection. Nano Energy 42, 300–306.
- 39. Yang, P.-K., Lin, L., Yi, F., Li, X., Pradel, K.C., Zi, Y., Wu, C.-I., He, J.-H., Zhang, Y., and Wang, Z.L. (2015). A Flexible, Stretchable and Shape-Adaptive Approach for Versatile Energy Conversion and Self-Powered Biomedical Monitoring. Adv. Mater. 27, 3817–3824.
- 40. Chen, C., Chen, L., Wu, Z., Guo, H., Yu, W., Du, Z., and Wang, Z.L. (2020). 3D double-faced interlock fabric triboelectric nanogenerator for bio-motion energy harvesting and as self-powered stretching and 3D tactile sensors. Mater. Today 32, 84–93.
- Zhang, H., Wang, H., Zhang, J., Zhang, Z., Yu, Y., Luo, J., and Dong, S. (2020). A novel rhombic-shaped paper-based triboelectric nanogenerator

CellPress



for harvesting energy from environmental vibration. Sens. Actuators, A $302,\,111806.$

- Subbaroyan, J., Martin, D.C., and Kipke, D.R. (2005). A finite-element model of the mechanical effects of implantable microelectrodes in the cerebral cortex. J. Neural. Eng. 2, 103–113.
- Jang, S., Shim, H., and Yu, C. (2022). Fully rubbery Schottky diode and integrated devices. Sci. Adv. 8, eade4284.
- 44. Xu, J., Wang, S., Wang, G.-J.N., Zhu, C., Luo, S., Jin, L., Gu, X., Chen, S., Feig, V.R., To, J.W.F., et al. (2017). Highly stretchable polymer semiconductor films through the nanoconfinement effect. Science 355, 59–64.
- Yamamura, A., Sakon, T., Takahira, K., Wakimoto, T., Sasaki, M., Okamoto, T., Watanabe, S., and Takeya, J. (2020). High-Speed Organic Single-Crystal Transistor Responding to Very High Frequency Band. Adv. Funct. Mater. 30, 1909501.
- Yamamura, A., Watanabe, S., Uno, M., Mitani, M., Mitsui, C., Tsurumi, J., Isahaya, N., Kanaoka, Y., Okamoto, T., and Takeya, J. (2018). Wafer-

scale, layer-controlled organic single crystals for high-speed circuit operation. Sci. Adv. 4, eaao5758.

- 47. Wang, W., Jiang, Y., Zhong, D., Zhang, Z., Choudhury, S., Lai, J.-C., Gong, H., Niu, S., Yan, X., Zheng, Y., et al. (2023). Neuromorphic sensorimotor loop embodied by monolithically integrated, low-voltage, soft e-skin. Science 380, 735–742.
- Koo, J.H., Kang, J., Lee, S., Song, J.-K., Choi, J., Yoon, J., Park, H.J., Sunwoo, S.-H., Kim, D.C., Nam, W., et al. (2023). A vacuum-deposited polymer dielectric for wafer-scale stretchable electronics. Nat. Electron. *6*, 137–145.
- 49. Matsuhisa, N., Niu, S., O'Neill, S.J.K., Kang, J., Ochiai, Y., Katsumata, T., Wu, H.-C., Ashizawa, M., Wang, G.-J.N., Zhong, D., et al. (2021). High-frequency and intrinsically stretchable polymer diodes. Nature 600, 246–252.
- Jiang, Y., Zhang, Z., Wang, Y.-X., Li, D., Coen, C.-T., Hwaun, E., Chen, G., Wu, H.C., Zhong, D., Niu, S., et al. (2022). Topological supramolecular network enabled high-conductivity, stretchable organic bioelectronics. Science 375, 1411–1417.