



RESEARCH ARTICLE

Thin, soft, garment-integrated triboelectric nanogenerators for energy harvesting and human machine interfaces

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Abstract

The applications of triboelectric nanogenerators (TENGs) in wearable electronics for energy harvesting and motion sensing have raised extensive attentions, since TENGs enable to convert body motions induced mechanical energy into electrical signals. The development of thin, soft, and garment-integrated TENGs would be an important solution for the power management in wearable electronics as well as self-powered sensors. Here, we report materials, device designs, processing routes for garment-integrated TENGs (G-TENGs) and demonstrations of the G-TENGs in wearable energy harvesting and human-machine interfaces. The G-TENGs adopt a simple layout with two soft silicone layers and one graphene-coated fabric layer, exhibiting great flexibility, air-permeability, and robust durability. Furthermore, the G-TENGs present outstanding electrical characteristics with open-circuit voltage and short-current outputs as great as 213.75 V and 3.11 μ A, under a constant frequency and stress of 3 Hz and 5.6 kPa, respectively. The excellent mechanical properties of the G-TENGs allow them tolerating toward over 1000 cycles of bending, stretching and twisting, and maintaining unchanged electrical outputs after these deformations. The stable electrical outputs and the excellent mechanical performance of the G-TENGs provide a high potential in self-powered sensors, energy harvesting, human-machine interfaces and many others.

KEYWORDS

garment-integrated electronics, graphene, human machine interfaces, textile electronics, triboelectric nanogenerators

1 | INTRODUCTION

Yiming Liu, Chunki Yiu, Huiling Jia, and Tszhung Wong contributed equally to this work.

Recently, wearable electronics have obtained extensive attention for their enormous potential in various fields,

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including healthcare monitoring,^{1–6} clinical medicine,⁷ and the human-machine interfaces.^{8–14} Developing flexible self-powered generators has become a trend for portable electronics, as which can continuously provide power support for wearable electronics.¹⁵ Recent advances in materials and electronic devices indicate various kinds of self-powered technologies, such as piezoelectric,^{16–19} triboelectric,^{20–26} electromagnetic,²⁷ and pyroelectric,²⁸ which can be used in flexible powering devices. Among these technologies, triboelectric nanogenerators (TENGs) have proven to be a great candidate as energy harvesting components for wearable electronics,^{15,29–32} due to their advantages of excellent electrical performance and mechanical properties. The working principle of TENGs is based on the coupling effect between triboelectrification and electrostatic induction in the functional layers via contact-separation modes or shearing modes, providing a highly feasible, effective, and practical approach for the realization of the conversion from mechanical energy into electricity.^{33–36} With the advantages of superior power density and stable electrical performance, TENGs have been widely used as energy harvesters.¹⁵

Among various kinds of TENGs, textile based TENGs have shown the potential for wearable energy harvesters due to their highly flexible and lightweight features.^{37–39} The textile based TENGs can serve as cloths to collect energies from daily human motions and thus powering the wearable electronics in a remarkable performance.^{40,41} Moreover, the soft and light weight nature of textile enable such kind of TENGs present excellent mechanical characteristics that can be compatible with our daily motions. One of the key materials in TENGs is the conducting materials, as which typically serve as electrodes. However, the majority of the reported ones are metals and conductive oxides, such as Cu,^{42,43} Au,⁴⁴ Ag,⁴⁵ and ZnO^{46–48} in fiber or nanoscale formats. In addition, the complicated processing routes of these materials into textile like formats increase the large-area fabrication cost, limiting their wide applications. Using carbonaceous fillers in textile-based TENGs, mostly as conducting materials, is another promising strategy.⁴⁸ As a typical carbonaceous material, graphene has proven to be an ideal electrode material because of its high electrical and thermal conductivity.^{49,50} Furthermore, the excellent electrostatic performance in graphene has been investigated and shown it is capable of storing electric charges and acting as a conductive material in TENGs.^{51–53} However, many reported graphene based TENG showed a low output power with a few voltages and currents, and the chemically grown graphene is only applicable to flat surfaces (Table S1). Therefore, the formation of graphene on uneven textile surface, as the

conductor of stretchable and durable TENGs, in a simple fabrication process and low cost is critical, and quite required to boost the output power for extending TENGs applications in wearable electronics.

Herein, we report a class of materials, device design, processing routes for garment-integrated triboelectric nanogenerators (G-TENGs), which offers a new strategy of high-throughput, large-area and air-permeable self-powered electronics for applications for energy harvesting and human machine interfaces. The G-TENGs adopt normal fabrics as the wearable precursor, a simple dyeing process of graphene for electrode coating, and quick blade coating of soft silicone for high throughput process. This kind of G-TENGs exploits advances in materials and mechanics, serving as the foundations for realizing soft, stretchable, ultrathin (thickness of 0.3 mm), and lightweight (50 mg cm⁻²) devices. Punching holes with a density of 9 cm⁻² enables the G-TENGs to exhibit good air-permeable characteristics (Figure S1). The excellent electrical negative nature of polydimethylsiloxane (PDMS) silicone allows the device yielding a high electrical output with the open-circuit (OC) voltage and short-circuit (SC) current of 213.75 V and 3.11 μ A at a stress of 5.6 kPa. Moreover, the electrical outputs of the G-TENGs show high sensitivity to tactile pressing, and thus allow it to serve as wearable sensors for human machine interfaces.

2 | MATERIALS AND METHODS

2.1 | Fabrication of GF-TENG

The fabrication started with a clean and soft nylon fabric with an overall size of 70 mm \times 70 mm \times 0.1 mm. Immersing the fabric into a container filled with graphene conductive paste (Nanjing Xianfeng Nanomaterial Technology Co., Ltd.) for 30 s allowed the fabric full fill with conducting materials. A glass sheet cleaned with acetone, ethanol, and deionized water (DI water), sequentially was used as substrate. The fabric was put onto the glass and then dried in an oven at a temperature of 100°C for 10 min. After drying, pouring PDMS precursor onto both sides of the graphene coated fabric, and then squeezing the redundant PDMS away by a self-developed blade coater, keeping it a pre-designed thickness. The sample was put in an oven at a constant temperature of 120°C for 2 min to cure completely. By placing the cured sample onto a flat foam board, a puncher with a needle density of 9 cm⁻² and pore size of 0.66 mm is applied to create arrayed holes on the sample for air breathing.

2.2 | Characterization

The open-circuit voltage and resistance were measured by a DAQ6510 data acquisition/multimeter system. The short-circuit current was calculated by measuring the voltage of a fixed value resistor which connected in series with the triboelectric nanogenerator (the voltage was measured by PL3516/P Powerlab 16/35, which owns much lower noise signal and higher sampling rate than the DAQ6510 multimeter system).

3 | RESULT AND DISCUSSION

Figure 1A presents a schematic illustration of an application scenario of G-TENGs, where a person wears a top cloth with several G-TENGs on it. The G-TENGs own a sandwich structure with a conductive graphene-coated nylon fabric sealed in between two thin PDMS layers (Figures 1B and S2). Figure 1C shows the scanning electron microscopy (SEM) images of the nylon fabric with

and without graphene, where the dense graphene attached on the fabric indicates the high efficiency of this coating method. A photo of a piece of G-TENG with its enlarged details are shown in Figure 1D. The overall dimension of the G-TENG is 70 mm × 70 mm × 0.3 mm (length × width × thickness). The ultrathin thickness and robust mechanical properties of the G-TENGs afford high flexibility, stretchability, and reversibility, that can endure different mechanical deformations, such as folding, stretching, and rolling (Figures 1E, S3, and S4). Figure 1F shows the working mechanisms of this G-TENG based on single-electrode mode, similar to other kinds of TENGs that are based on triboelectrification effect and electrostatic induction.¹⁵ External applied force enables contact happening between the surface of PDMS and the touching object, and therefore induces charge transfers. Separation of the PDMS and touching object leads to an electric potential difference form in between them, resulting in an instantaneous generated electrical outputs. The charges are in equilibrium when the two materials are completely separated. Thus, a continuous

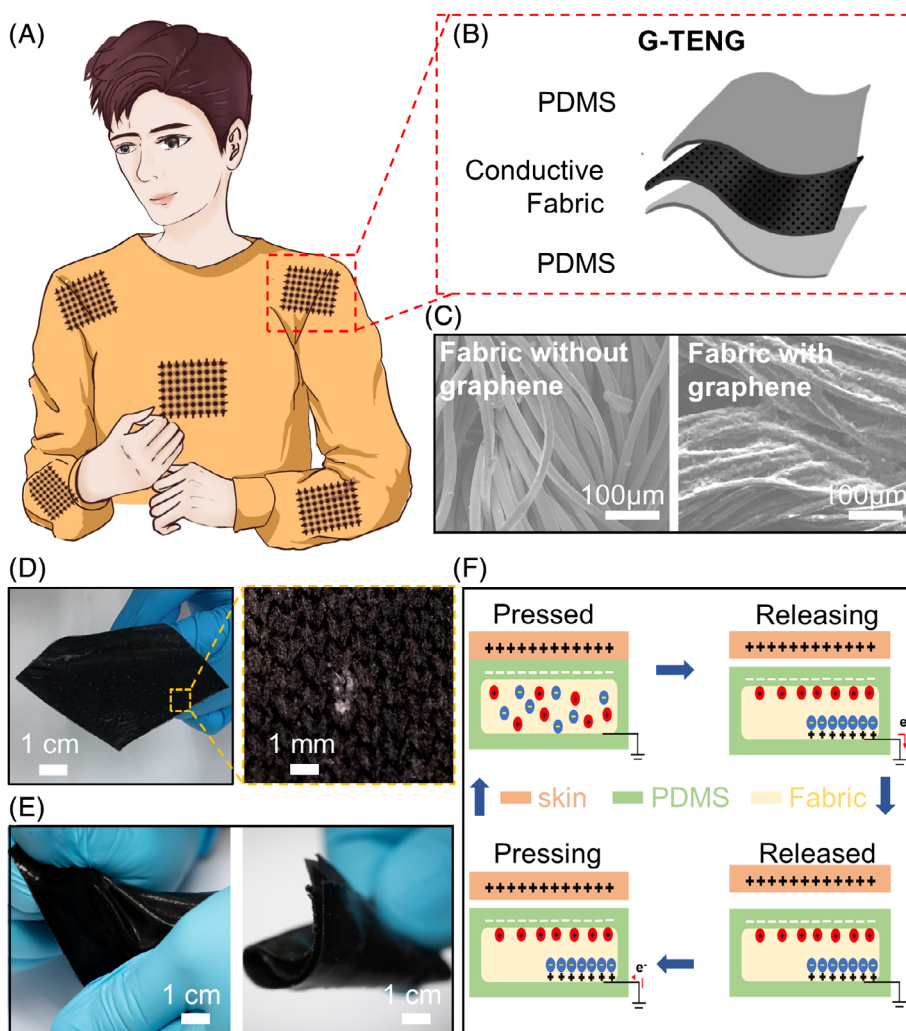


FIGURE 1 Design and working principle of the air permeability G-TENGs. (A) A cartoon illustration of a person wearing G-TENG sewn clothes. (B) The schematic diagram of the G-TENG. (C) The scanning electron microscopies of the fabric without and with graphene. (D) Optical images of the flexible G-TENG with enlarged details. (E) Optical images of the G-TENG under two mechanical deformations, including stretching, and folding. (F) Illustration of working principles of the G-TENG

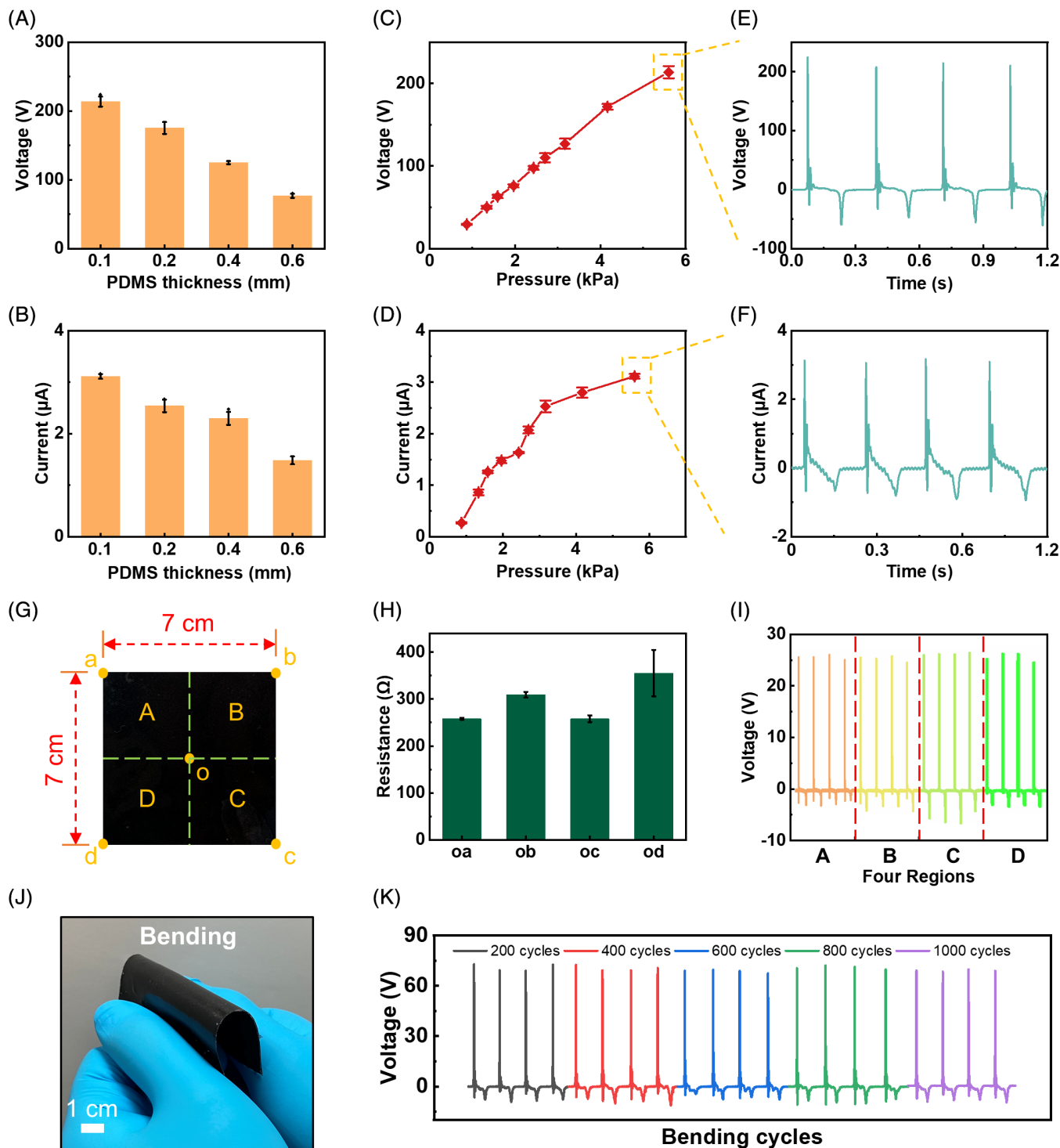


FIGURE 2 The electrical characteristics of the G-TENG. The open-circuit voltage (A) and short-circuit current (B) in G-TENG as a function of triboelectric PDMS layer thickness (weight ratio of crosslink: PDMS = 1:10). The open-circuit voltage (C, E) and short-circuit current (D, F) in the G-TENG with PDMS thickness of 0.1 mm as a function of stress at a constant frequency of 3 Hz. (G) Optical image of the G-TENG and the conductivity measurement regions “a,” “b,” “c,” and “d.” The resistances between the origin “O” and the four vertices of the G-TENG (H) and open-circuit voltages (I) by finger tapping in the four regions with a constant stress and frequency of 14.8 kPa and 3 Hz. (J, K) The optical image of G-TENG under continuous bending by hand and the open-circuit voltage of GF-TENG under a constant stress and frequency of 1.6 kPa and 3 Hz after 1000 bending cycles at a radii of 0.75 cm

output current could be derived from such repeated contact-separation motions. The schematic illustration of the fabrication process of the G-TENG is shown in Figure S5. First, a nylon fabric is immersed into an aqueous solution containing graphene to coat the conducting graphene uniformly. Then, pouring PDMS on the graphene dyed fabric and squeezing the excess PDMS away (Figure S6) by a customized blade coater form the encapsulation layers on both sides of the fabric. Punching the PDMS coated fabric with a multineedle puncher (pore diameter of 0.66 mm, density of 9 cm^{-2}) offers the G-TENGs great air-permeability. Figure S1 presents the results of measurements of the water vapor transmission rate (WVTR) of the G-TENG, and it is found that the G-TENG is air-permeable with the WVTR value of $0.35 \text{ gh}^{-1} \text{ m}^{-2}$ after 24 h, comparable with the air-breathing commercial nylon fabric (WVTR, $0.51 \text{ gh}^{-1} \text{ m}^{-2}$).

The thickness of the triboelectric layers in TENGs is one of the key parameters determining the electrical performance. To explore the optimized thickness of the tribo-material and its influence on the electrical output performance of the G-TENGs, a series devices with different thickness of PDMS layers in the G-TENGs ranging from 0.1 to 0.6 mm were investigated. Figures 2A,B and S7 summarize the OC voltage and the SC current in G-TENGs with different PDMS thicknesses, where we can observe the both OC voltage and SC current of G-TENGs significantly increase with the deduction of the PDMS thickness. Under a constant testing pressure of 5.6 kPa, the maximum OC voltage and SC current are 213.75 V and $3.11 \mu\text{A}$ for G-TENGs with a PDMS thickness of 0.1 mm, indicating that a thinner thickness of the PDMS layer could improve the electrical performance of the device. However, textile based TENGs with thinner triboelectric layers would lead to a high damage risk as they tend to continuously contact and separate with human skin for mechanical energy harvesting.⁵⁴ Herein, we use 0.1 mm thick PDMS film as the triboelectric layers for the G-TENGs to maintain the robust durability without further decreasing the triboelectric layer. Figure 2C,D show the OC voltage (Figure 2C) and SC current (Figure 2D) of the device as functions of different applied pressures, ranging from 0.87 to 5.6 kPa, at a frequency of 3 Hz. The maximum electrical responses at 5.6 kPa are 213.75 V and $3.11 \mu\text{A}$ (charge transfer, $0.9 \mu\text{C}$; more details in Figure S8 and Part I Surface charge transfer calculation), respectively, and the detailed electrical outputs versus time show the great output stabilities (Figure 2E,F). Figure S9 shows the OC voltage of the G-TENG with three stretching rates (0%, $\sim 20\%$, and $\sim 30\%$) under a constant stress and frequency of

2.6 kPa and 3 Hz, and it is found that the stretching condition has no obvious influence on the electrical signal output of the G-TENG. Figure S10 presents three different testing conditions as human skin interfaces with the G-TENG, including G-TENG without holes on surface under dry hand tapping, G-TENG with holes on surface under dry hand tapping, and G-TENG with holes on surface under wet hand tapping, that yield OC voltage of 28.9, 28.1, and 19.3 V, respectively at a constant stress and frequency of 0.87 kPa and 3 Hz. It can be found that humidity in human skin has a negative effect on electrical performance of the G-TENG, which we believe future works on materials and designs can conquer this problem.

The high throughput processing route of enables the graphene coated fabric exhibiting very uniform conductivity over the entire working region, and therein build a very strong foundation for G-TENGs with great uniformity in performance. A representative graphene coated fabric with demission of $7 \text{ cm} \times 7 \text{ cm}$ was used to identify the uniformity via measuring the conductivity at different areas (Figure 2G), that associating with the measurement of resistance between the center “o” and the four vertexes (“a,” “b,” “c,” and “d”). Figure 2H shows the resistances of these four areas, that equal to 258, 309.4, 258, and 355.4Ω in average, respectively. The OC voltages of the resulted G-TENG were measured by finger tapping in the four regions with constant stress and frequency of 14.8 kPa and 3 Hz, respectively. Very stable voltage outputs of $\sim 25 \text{ V}$ can be obtained from four different areas, indicating the great uniformity of the G-TENGs and potential in large area applications (Figure 2I). Great mechanical durability and deformability are critical for wearable textile devices under continuous large deformations such as bending and twisting. To investigate the durability of the G-TENGs under intensive mechanical deformations, we study its long-term electrical performance by testing it under thousands of bending cycles by hand. Notably, after 1000 times bending cycles at a radii of 0.75 cm, there is no apparent difference in the voltage output of the G-TENGs (Figure 2J,K). The outstanding mechanical flexibility and great uniformity of the G-TENGs allow them serving as self-powered/energy harvesting devices based on daily human activities.

Figure 3A demonstrates one practical application of the G-TENGs in human machine interfaces, where five $1.5 \text{ cm} \times 1.5 \text{ cm}$ G-TENG units integrated on the sleeve of a cloth serve as self-powered sensors to remotely control a small tele-car. The control system of the human machine interface adopts “Arduino” as the micro-

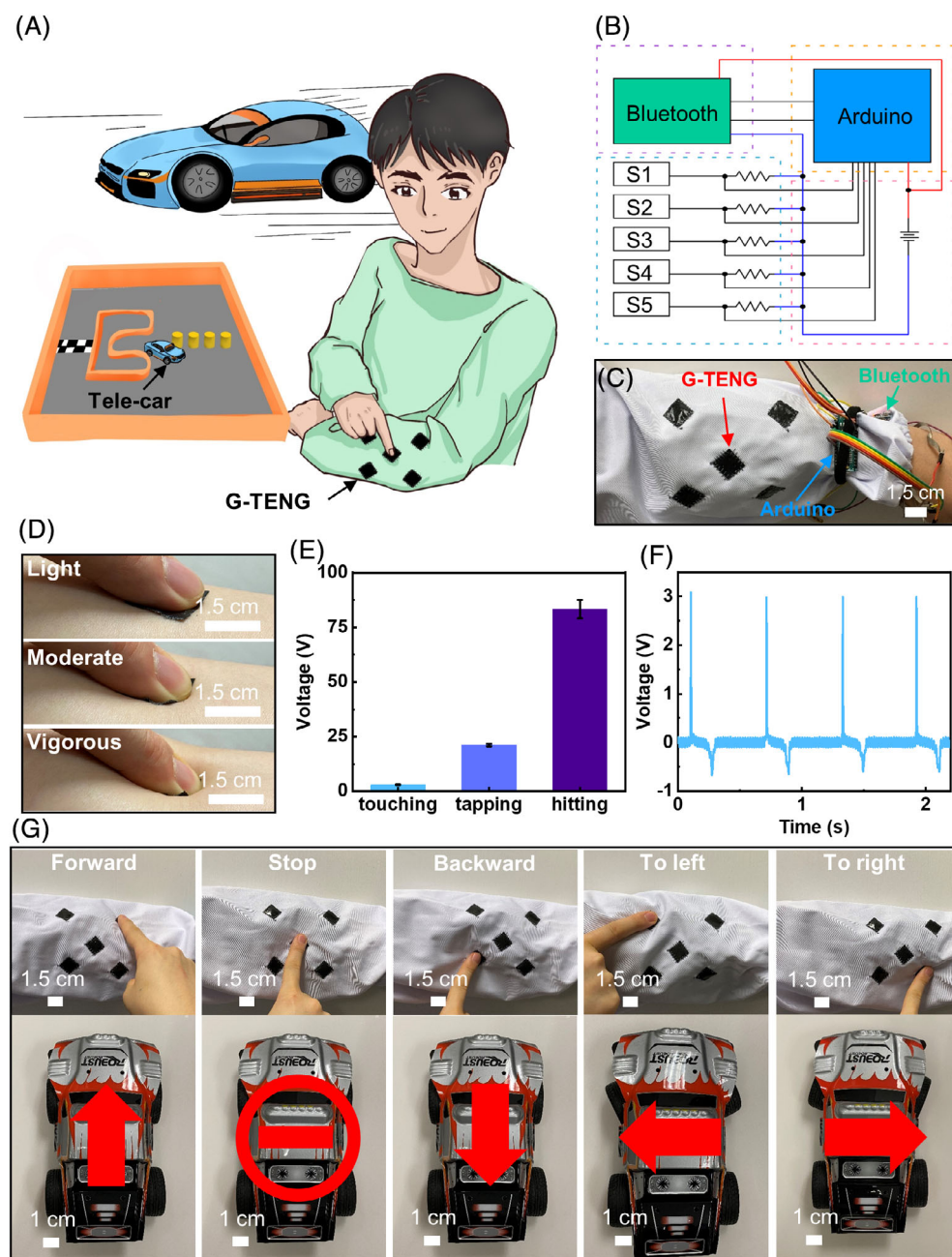


FIGURE 3 Demonstration of G-TENGs in human machine interfaces. (A) Schematic diagram of remotely controlling tele-car through G-TENG. (B) The schematic diagram of the testing circuit. (C) Optical image of the whole controlling system mounted onto human clothes. (D) Optical images illustrating three different external loads of light tapping, middle tapping, and hard tapping on a G-TENG. (E, F) Electrical responses in the G-TENG sensors to the three kinds of touching loads. (G) Photos of controlling process of the tele car, where fingers tapping on different buttons correspond to “forward,” “stop,” “backward,” “left,” and “right”

controller for collecting the sensing data from the five G-TENG units (working as pressure sensors), then the Arduino board (Arduino MEGA 2560, ARDUINO) sends the data to the coupled control system mounted on the tele-car for further analysis by a Bluetooth (WH-BLE103) module (Figure 3B). The whole system is powered by a commercial Li battery (60 mAh), which can support 4 h operating time. The wireless transmission module is mounted onto the user's sleeve nearing the G-TENGs based sensing system (Figure 3C). The largest communication distance between the control system and the tele-car can reach up to 3.84 m. Different electrical responses from different tapping force in 5 G-TENG sensors serve

as the commands to control power outputs in the tele-car that are directly relevant to the speed. The outputs to the tele-car are set at three levels of “light (~ 1.5 cm/s),” “moderate (~ 4.5 cm/s),” and “vigorous (~ 7.4 cm/s)” that are defined by different ranges of the tapping force applied by a finger to the G-TENGs associating to light (≤ 3.6 kPa), middle ($3.6 \sim 18.2$ kPa) and hard ($18.2 \sim 74.7$ kPa) (Figure 3D). Figure 3E presents the OC voltage of the $1.5 \text{ cm} \times 1.5 \text{ cm}$ G-TENG under the three different pressure triggered by finger tapping. Different forces applied on the G-TENGs resulting in corresponding voltage outputs serve as the analog signals and then be converted into digital signals by the micro

controller to control the velocity of the tele-car. The stable electrical performance of the G-TENGs offers repeatable and precise control behaviors in the human machine interface (Figure 3F). The user can control the tele-car to drive in four different directions and stop by pressing the corresponding G-TENGs on the sleeve (Figure 3F and Movie S1). Furthermore, the driving velocity could be changed by adjusting the pressing strengths. The high sensitivity toward human motions and the smooth control of a tele-car indicate that the G-TENG devices own a high potential in the human-machine interfaces.

The ability to harvest energy from human motions makes the G-TENG to be a desirable portable power

source. To further explore the power supply performance of the device, an energy harvesting cloth is demonstrated by sewing four G-TENGs at the locations of forearm, shoulder, tummy, and back (Figure 4A,B), and the electrical outputs are collected by contacting and separating from human skin during human different daily activities, such as walking, running, and jumping (Figure 4C). Both the OC voltage and SC current are strongly relevant to the types and intensities of the corresponding activities. No doubt, intense activity, such as jumping, can generate the greatest voltage values. Taking the G-TENG mounted on the tummy area (A3) as an example, the OC voltage and SC current show quite different values under walking, running

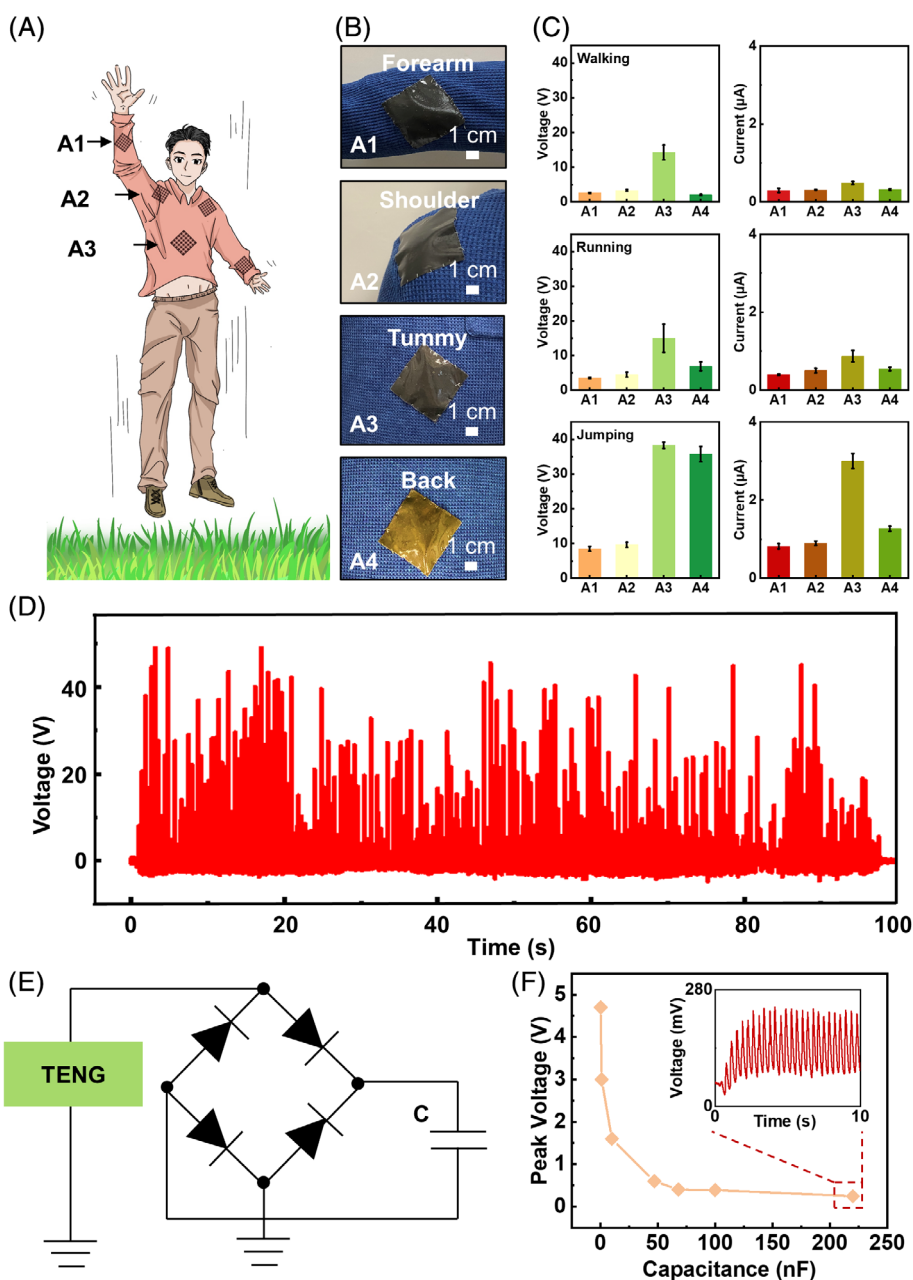


FIGURE 4 Demonstration of G-TENGs in wearable energy harvesting. (A) Cartoon illustration of a person wearing G-TENG integrated clothes for energy harvesting from daily activities. (B) The optical images of G-TENGs integrated onto four parts of clothes, including the area of forearm, shoulder, tummy, and back. (C) The open-circuit voltage and short-circuit current of the G-TENGs at different areas response to three different activities (walking, running, and jumping). (D) Electrical response of the G-TENG at tummy area to jumping at a frequency of 1 Hz for 100 s. (E) The charging circuit design for G-TENG, where a rectifying bridge is used for covert electrical outputs into DC inputs, and the capacitor is used for collecting charges. (F) The peak voltage of various capacitors can be charged by the G-TENG mounted on tummy area, under jumping activity at frequency of 1 Hz

and jumping, corresponding to 14.28 V@0.49 μ A, 15 V@0.87 μ A, 38.28 V@3.16 μ A, respectively (Figure 4C). This energy harvesting cloth is capable of continuous generating electricity during activities (Figure 4D). To store the harvested energy, capacitors are introduced to collect electrical outputs with the assistance of a rectifying bridge circuit (Figure 4E), which helps regulate the alternating current (AC) to the direct-current (DC) output. As shown in Figure 4F, the peak voltage of the G-TENG drops from 4.7 to 0.245 V along with an increasing capacitance from 0.22 to 220 nF. In addition, the G-TENG can also output energy for instant applications, and corresponding proof of concept is shown in lightning light emitting diodes (LEDs). Finger tapping one G-TENG can light over 80 green LEDs, as shown in Movie S2. The stable and persistent electrical outputs of the G-TENGs demonstrate the potential of high feasibility of daily, wearable energy suppliers.

4 | CONCLUSION

In summary, an easy-fabricated, ultrathin, wearable, and flexible garment integrated energy harvesters is introduced in this work via combining the advances of materials science and device designs in TENGs. The simple processing route by washing normal fabrics in graphene-based solutions and one step sealing by coating with PDMS in G-TENGs offers a promising method for large area and high throughput wearable and self-powered electronics. The materials and geometries optimization of the G-TENGs allow the device exhibiting great energy output efficiencies and excellent mechanical properties. Furthermore, the G-TENGs could be easily sewn on clothes and generate considerable electrical signals during daily human motions like walking, jumping, or running, and used as both energy harvester and self-powered sensors. Demonstrations of such a flexible and robust device in pressure sensing and energy harvesting from body movements provide a promising route to practical applications of self-charging portable electronics and the human-machine interfaces.

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CONFLICT OF INTEREST

The authors declare that there is no conflict of interest regarding the publication of this article.

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SUPPORTING INFORMATION

Additional supporting information may be found online in the Supporting Information section at the end of this article.

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