# A Controlled Biodegradable Triboelectric Nanogenerator Based on **PEGDA/Laponite Hydrogels**

Zhe Li, Cong Li, Wei Sun, Yuan Bai, Zhou Li,\* and Yulin Deng\*

Cite This: ACS Appl. Mater. Interfaces 2023, 15, 12787-12796



ACCESS	III Metrics & More	🖭 Article Recomm	nendations	s Supportin	g Information
<b>ABSTRACT:</b> Implantable and wearable transient electronics based on papogenerators have been applied in self-powered sensing electrical-			H H THE PEGDA		et et et

rs have been applied in self-powered sensing, electrical stimulation therapy, and other fields. However, the existing devices have a poor ability to match with the shapes of human tissues, and the degradation processes cannot meet individual needs. In this work, a PEGDA/Lap nanocomposite hydrogel was prepared that was based on biocompatible polyglycol diacrylate (PEGDA) and laponite, and a biodegradable single-electrode triboelectric nanogenerator (BS-TENG) was built. The PEGDA/Lap hydrogel has enhanced flexibility and mechanical and electrical performance. Its strain was 1001.8%, and the resistance was 10.8. The composite hydrogel had a good biocompatibility and could effectively promote the adhesion of cells. The BS-TENG could be used as a self-powered device to light an LED and serve as an active



sensor for real-time monitoring of breath and various human movements. More importantly, the device could be degraded controllably without any harm. Therefore, BS-TENGs will be mainstream in diagnosis and treatment and play an important role in biomedical science.

**KEYWORDS:** hydrogel, biodegradable, controllable, self-powered, TENG

# 1. INTRODUCTION

In recent years, wearable devices, medical implantable devices, electronic skin, and intelligent electronic fabrics have been proposed.<sup>1-4</sup> The applications of flexible electronics combined with human organs and tissues have been more and more extensive.<sup>5</sup> The rapid development of flexible electronics and circuits will promote efficient communications between people and information.<sup>6-9</sup> However, most of the traditional electronics have rigid structures and limited energy supplies,<sup>10</sup> and they easily cause environmental pollution. There are still shortcomings in the aspects of conformal integration with tissues, microsignal recognition, and long-term stability. With the improvement of the medical level, the requirements for portable, highly sensitive, self-powered, side-effect-free embedding of electronics will become higher. In addition, based on thinking from humans on the growing energy and environmental issues, recyclability and degradability have also become the key parameters for device.<sup>11,12</sup>

Triboelectric nanogenerator (TENG) is a novel technology that can realize self-powered sensing and low-frequency energy harvesting, which can meet the long-term energy requirements of electronics and has great potential in flexible wearable, and implantable electronics.<sup>13–16</sup> In addition, in order to avoid the removal and replacement of implanted medical electronics, a variety of biodegradable TENGs (BD-TENGs) have been developed and applied in electrical stimulation therapy, physiological signal monitoring, and other fields.<sup>17-19</sup> However, the selection of flexible materials for BD-TENGs is limited, the tensile capacity needs to be improved,  $^{20-22}$  and the degradation times of the devices cannot meet the personalized requirements. Therefore, it is urgent to explore high-performance flexible stretchable materials, and construct controllable biodegradable TENGs to meet the various human needs for medical electronics.

Compared with traditional metal electrodes, hydrogels have good flexibility, stretchability, and self-healing activity.<sup>23-26</sup> However, the improvement of conductivity,<sup>27,28</sup> mechanical property,<sup>29,30</sup> biocompatibility, or biodegradability,<sup>31,32</sup> and the response to external stimuli will promote their practical applications.<sup>25,33</sup> Polyethylene glycol (ethylene glycol) diacrylate (PEGDA) hydrogels have been used as new scaffolds in tissue engineering and regenerative medicine due to their highwater contents, biosafety, and simple preparation processes.<sup>34,35</sup> In previous studies, it showed that the introduction of inorganic clay nanoparticles (Laponite XLS) and conductive

Received: December 12, 2022 Accepted: February 23, 2023 Published: March 1, 2023







Figure 1. Schematic illustration of the preparation process of BS-TENGs based on the multifunctional PEGDA/Lap hydrogel and their applications in monitoring physiological signals.

ions can improve the overall mechanical and electrical properties of the chemically cross-linked PEGDA hydrogel system.<sup>36</sup> The prepared nanocomposite hydrogels have enhanced extensibility, toughness, tensile strength, and high conductivity.

Herein, PEGDA and Laponite nanocomposites (Lap) were applied as raw materials to prepare PEGDA/Laponite (PEGDA/Lap) composite hydrogels. A flexible biodegradable single-electrode TENG (BS-TENG) was also assembled. When the amount of Laponite was 10 wt %, the composite hydrogel has the optimal electrical and mechanical properties. The strain of the PEGDA/Lap composite hydrogel is about 1001.8%, and the resistance is about 10.8 $\Omega$ . The material has a good biocompatibility and can promote cell adhesion. The developed BS-TENG with the size of 25 mm  $\times$  15 mm has an open-circuit voltage  $(V_{OC})$  of about 10.4 V and can work stably for 10<sup>6</sup> cycles. BS-TENGs can not only supply electricity to commercial electronics by harvesting mechanical energy but also serve as a self-powered sensor to detect breathing, joint motion, and other physiological indicators of the human body. In addition, the device can degrade within 90 days, and the degradation can be controlled by temperature and lipase. In conclusion, the flexible BS-TENG prepared from PEGDA/Lap composite hydrogels has good potential in energy harvesting, self-powered wearable electronics, and biomedical sensors.

## 2. MATERIALS AND METHODS

**2.1. Materials.** Polyethylene glycol ( $M_w$ : 10 000), lithium phenyl-2,4,6-trimethylbenzoylphosphinate (LAP), sodium chloride (NaCl), and 10× PBS buffer solutions were purchased from Aladdin Reagent Co. Laponite XLS ( $[Mg_{5.34}Li_{0.66}Si_8O_{20}(OH)_4]$  Na<sub>0.66</sub> (92.32 wt %) + Na<sub>4</sub>P<sub>2</sub>O<sub>7</sub> (7.68 wt %) was obtained from Wentianqihang (Jinan) Technology Innovation and Development Co., Ltd. Cell culture supplies including standard Dulbecco's modified Eagle's medium (DMEM, Gibco), penicillin/streptomycin (Gibco), and fetal bovine serum (FBS, Gibco) were purchased from Beijing Solarbio Science & Technology Co., Ltd. L929 fibroblasts were obtained from Beijing Institute of Nanotechnology and Nanosystems. L929 cells were

cultured in medium containing high glucose DMEM, 10% FBS, 1% penicillin, and streptomycin.

**2.2. Preparation of the Hydrogel.** The hydrogel was prepared by a one-pot method. Specifically, a certain amount (5, 10, 20 wt %) of Laponite XLS ( $[Mg_{5.34}Li_{0.66}-Si_8O_{20} (OH)_4]Na_{0.66} (92.32 wt %) + Na_4P_2O_7 (7.68 wt %))$  was added into deionized water. The solution was dissolved by stirring thoroughly and then left at room temperature (~25 °C) for at least 20 min to obtain the Laponite XLS stock solution. PEGDA solids ( $M_w = 10\,000$ , final concentration of 10 wt %), 0.5% (w/v) photoinitiator (LAP) were introduced inside, dissolved by stirring fully, and dispersed by ultrasonication to form a homogeneous solution. After being gelatinized in a glass mold, the mixture was exposed to 405 nm light at 10 nm cm<sup>-2</sup> intensity for 2 min, and the PEGDA/Lap hydrogel was prepared.

**2.3. Material Properties.** Tensile tests of hydrogels were performed on an ESM301/Mark-10 system (Mark-10 Company, New York, NY, USA) at a tensile rate of 10 mm min<sup>-1</sup>. The size of the tensile test sample was  $20 \times 10 \times 1$  mm. Fourier transform infrared (FTIR) spectra were obtained by using a VERTEX80v spectrometer (Bruker, Karlsruhe, Germany). All spectra were scanned from 4000 to 400 cm<sup>-1</sup>. The morphology and the energy-dispersive spectroscopy (EDS) mapping of the samples were obtained by using a cold field scanning electron microscope (SEM, SU8020, HITACHI, Japan).

**2.4. Electrochemical Measurement.** Electrochemical spectroscopy (EIS) of hydrogels was performed on an electrochemical workstation (CHI 650E, Chenhua, Shanghai). EIS was performed in the open circuit range from 0.1 Hz to 1 MHz. A linear motor (LinMot E1100) was used to provide a periodic contact separation motion for the electrical measurement of a single-electrode nanogenerator. Keithley 6517 electrometer was applied to measure the changes of open-circuit voltage ( $V_{oc}$ ), short-circuit current ( $I_{sc}$ ), and transfer charge ( $Q_{sc}$ ) of hydrogels under different pressures and deformations in real time. An oscilloscope (LeCroy HDO6104) was used to collect and record the data. In the measurement of physiological signals, breathing and other inductive signals were tested by the oscilloscope directly, and an electrometer was supplied to detect finger bending, sliding, tapping, and other signals.

**2.5. Cell Viability Test.** Hydrogels with different ratios of PEGDA and Laponite XLS were irradiated for 30 min under UV light for sterilization. They were cut into circles with a diameter of 14 mm and placed in a 96-well plate. L929 cells ( $2 \times 10^4$  per well) were seeded on hydrogels and cultured with DMEM supplemented with 1%



Figure 2. Characterization and properties of PEGDA hydrogels with different Laponite doping. (A) The design principle of the PEGDA/Lap hydrogel. (B) SEM images. (C) FTIR spectra. (D) The electrochemical impedance spectra (EIS). (E) Tensile strain-stress curves. (F) Cell viability of PEGDA and PEGDA/Lap hydrogels.

penicillin/streptomycin and 10% FBS. The cells were incubated for 1, 2, and 3 days at 37 °C in a humid environment containing 5% CO<sub>2</sub>, respectively. The viability and proliferation of L929 cells were detected by a Cell Count Kit-8 (CCK-8) assay. The medium was changed every day. After 1, 2, and 3 days of coculture, 10  $\mu$ L of CCK-8 solution was added to each well and incubated for 4 h at 37 °C in a humid environment with 5% CO<sub>2</sub>. Optical density values were measured at a wavelength of 450 nm using a multimode microplate reader (ThermoFisher Multiskan FC, USA). Three parallel control wells were obtained from each group.

**2.6. Cell Morphology and Immunofluorescence Staining.** After the hydrogel was treated as described above, it was cut into  $20 \times 20$  mm squares and put into a six-well plate. L929 cells ( $1 \times 10^5$  per well) were seeded on hydrogels and cultured in DMEM complete medium at 37 °C in 5% CO<sub>2</sub>. After 1, 2, and 3 days, the medium was removed, and the L929 cells on hydrogels were washed with 1× PBS buffer for three times and stained, respectively. They were stained with 1,1-dioctadecyl-3,3,3,3-tetramethylindocarbocyanine perchlorate (Dil) and 4',6-diamidino-2-phenylindole (DAPI) for 30 and 5 min, respectively. Immunofluorescence images were taken with a Leica

confocal fluorescence microscope (LECIA TCS SP8) at 488 and 565 nm emission filters.

**2.7. Degradation Assays.** To investigate the dissolution process and kinetics of the device, a series of dissolution assays were performed in 10 mL of  $1 \times$  PBS buffer at room temperature (25 °C) and 37 °C. In addition, the devices were placed in 10 mL of  $1 \times$  PBS buffer with 0.01 wt % lipase at room temperature (25 °C). Physical images of the device were taken periodically during the degradation.

# 3. RESULTS

**3.1. Preparation of PEGDA/Lap Composite Hydrogel.** Previous works have focused on the preparation of Laponite composite hydrogels with PEG of high molecular weight, but there have been few studies on whether Laponite nanoparticles could interact with low-molecular-weight PEG macromolecules in a similar manner to form hydrogels. In this study, PEGDA/Lap hydrogels were prepared according to the following experimental procedure (Figure 1).

PEGDA oligomers with a molecular weight of 10k were selected, and the final ratio in deionized water was 10 wt %.



**Figure 3.** Biodegradable single-electrode TENGs. (A) Schematic illustration of a BS-TENG based on PEGDA/Lap hydrogel. (B) The photos showed the BS-TENG has good flexibility and stretchability. (C) The mechanism of the BS-TENG in contact-separation single-electrode mode with skin. (D–F) The  $V_{oc}$ ,  $I_{sc}$  and  $Q_{sc}$  of the BS-TENG at a 1 Hz relative motion to the skin. (G) The simulated potential distributions for BS-TENG by the finite element method.

The interaction between Laponite nanoparticles and PEGDA polymer chains was assessed by varying the amount of Laponite (5, 10, and 20 wt %). As shown in Figure 2A, the hydrogel has a triple network structure. (i) PEGDA oligomers were chemically cross-linked to form a network. (ii) The PEG chains with high molecular weight had strong secondary interactions with Laponite nanoparticles, which enhanced the mechanical properties of the physical cross-linked hydrogel. (iii) The formation of the third network was attributed to the photo-cross-linking between the photoinitiator and the acrylate group on PEGDA. The synthetic hydrogels were studied by various characterization methods. SEM results showed that the low-molecular-weight poly(ethylene glycol) oligomer could form a porous hydrogel by interacting with Laponite nanoparticles in water (Figure 2B). The synthetic hydrogels were studied by various characterization methods. SEM results showed that the low-molecular-weight polyethylene glycol oligomer could form a porous hydrogel by interacting with Laponite nanoparticles in water (Figure 2B). From the EDS element mapping (Figure S1), it can be seen that Lap nanoparticles were evenly distributed in the entire PEGDA/ Lap hydrogel. The concentration of Laponite was closely related to its formation. The formation of PEGDA/Lap hydrogel was further characterized by FTIR (Figure 2C). According to the infrared spectrum of the freeze-dried hydrogel, 1466 cm<sup>-1</sup> was the C-H bending vibration peak of PEGDA. 1349, 1280, and 1249  $\text{cm}^{-1}$  were the  $\hat{O}-H$ bending vibration and C-H stretching vibration of PEGDA. The 1640, 980, and 650 cm<sup>-1</sup> were characteristic peaks of Laponite nanoparticles. The structure confirmed the existence of PEGDA and Laponite groups in the hydrogel.

**3.2. Physical Properties Characterization of PEGDA/ Lap Composite Hydrogels.** The conductive properties of the PEGDA/Lap composite hydrogels were also studied. Electrochemical impedance spectroscopy (EIS, Figure 2D and Figure S1A) showed that the equivalent resistance (ER, calculated by intersecting the *X*-axis) was decreased by the doping of Laponite, reaching a minimum value of  $10.8\Omega$  when the ratio of PEGDA and Laponite was 1:1. The results indicated that the introduction of an appropriate amount of Laponite nanoparticles could reduce the resistance and improve the conductivity of the composite hydrogel.

After the gelling ability of low-molecular-weight PEG oligomers and Laponite nanoparticles was verified, the physical properties of the PEGDA/Lap composite hydrogels were characterized. The effect of Lap's contents on mechanical properties of PEGDA hydrogel was investigated by compression (Figure S1B) and tensile tests (Figure 2E). Compared with pure PEGDA hydrogels, PEGDA/Lap hydrogels had 11.2 times the tensile resistance and were less prone to rupture under pressure, which proved that the addition of nano-Laponite significantly improved the overall mechanical properties of PEGDA hydrogels.

Cell viability is an important index to evaluate the biocompatibility of biomaterials. Polyethylene glycol has been shown to have good biocompatibility. To understand the effects of PEGDA/Lap hydrogels on cells, the adhesion and proliferation of L929 cells on PEGDA/Lap hydrogels containing different concentrations of Laponite nanoparticles (0, 5, 10, and 20 wt %) were evaluated by immunofluorescence staining images (Figure S3) and statistical analysis results (Figure 2F). The results showed that, compared with the PEGDA hydrogel, the PEGDA/Lap hydrogel doped with 10 wt % Laponite showed better cell activity and ability to promote cell proliferation. However, the biocompatibility of PEGDA/Lap decreased with 20 wt % Laponite doping. These results indicated that the introduction of appropriate amounts of Laponite nanoparticles could promote cell adhesion and proliferation. Therefore, considering all aspects, the hydrogel containing 10 wt % Lap was selected for subsequent experiments.

**3.3. Preparation and Output Characterization of BS-TENGs.** Considering the human tissue modulus (about 0.1–100 kPa) and the conformal integration between devices and tissues, we proposed a flexible and ultrasensitive biodegradable BS-TENG (BS-TENG) based on the excellent PEGDA/Lap hydrogels (Figure 3A). The BS-TENG was flexible and stretchable, as shown in Figure 3B. The PEGDA/Lap hydrogel

www.acsami.org



**Figure 4.** Demonstration of the BS-TENG for mechanical energy harvesting. (A) Dependence of the  $V_{oc}$  (blue),  $I_{sc}$  (green), and (B) power density on different external load resistances. (C) Circuit diagram for driving electronics with a rectifier and a capacitor. The charging curve of the BS-TENG at (D) different capacitance capacities (1–10  $\mu$ F) and at (E) different operating frequencies (1–5 Hz) for a 10  $\mu$ F capacitor. (F) Digital images of the BS-TENG powering a red-light-emitting diode (LED).

served as the electrode layer and triboelectric layer. An external conductive molybdenum (Mo) wire was supplied for the conductor and connected with the PEGDA/Lap hydrogel. A piece of Ecoflex film with the 25 mm  $\times$  15 mm was placed under the PEGDA/Lap hydrogel to insulate the lower surface of the BS-TENG from other objects. In the initial state (Figure 3C), the skin was in full contact with the BS-TENG, and the PEGDA/Lap hydrogel deformed under an external force. Equal positive and negative charges were generated on PEGDA/Lap hydrogel and skin, respectively. When the force was withdrawn, the skin and TENG gradually separated. The binding ability of the negative charges on the skin to the positive charges on the PEGDA/Lap hydrogel became weaker, resulting in the current flowed from the hydrogel to the external loop. Then, when the skin moved far enough, the charge movement stopped in the circuit. This was due to the positive charges on the PEGDA/Lap hydrogel completely escaping to the outside. Finally, when the skin was gradually close to the PEGDA/Lap hydrogel, the binding ability of the negative charges on the skin to the positive charges in the PEGDA/Lap hydrogel was enhanced. The potential difference between the skin and PEGDA/Lap hydrogel was reduced, and the current flowed from the ground to the hydrogel. When the PEGDA/Lap hydrogel was in full contact with the skin, the working mode returned to (i) and the cycle continued. The open circuit voltage  $(V_{oc})$ , short circuit current  $(I_{sc})$ , and short circuit transfer charge  $(Q_{sc})$  of the device were tested by an electrometer and oscilloscope.  $V_{oc}$ ,  $I_{sc}$  and  $Q_{sc}$  were 11.2 V, 1.9  $\mu$ A, and 11.7 nC, respectively (Figure 3D-F). The electrical potential on the skin and PEGDA/Lap hydrogel was also simulated by COMSOL software, and the results were consistent with the test results (Figure 3G).

The output voltage, current, or transferred charges can be affected by factors such as working frequency, working distance and device sizes. As shown in Figure S4, the  $V_{oc}$  of BS-TENGs at different frequencies and working distances was tested using a linear motor and an electrometer. A PTFE film with the size of 20 × 10 mm was fixed on the linear motor for the friction with the BS-TENG, and the working distance of the linear

motor was set as 20 mm. When the operating frequency changed from 0.5 to 100 Hz, the  $V_{\rm oc}$  was basically unchanged (Figure S4A, B). This indicated that the  $V_{\rm oc}$  of BS-TENG is not affected by the operating frequency. According to formula 1, the maximum  $V_{\rm oc}$  has a first-order linear relationship with x(t).<sup>37–39</sup> Although the frequency changed, the materials and the distance between the two triboelectric layers (working distance) in the system are constant. Therefore, the  $V_{\rm oc}$  was constant, which was independent of frequency. However, in the actual test, the  $V_{\rm oc}$  had a small range of changes due to air flow, electromagnetic wave interference in the environment, and other factors.<sup>40,41</sup>

$$U = \frac{\sigma x(t)}{\varepsilon_0} \tag{1}$$

Where *U* is the voltage between the two electrodes,  $\sigma$  is the triboelectric charge density, *x* is the distance between the two triboelectric layers, and  $\varepsilon_0$  is the dielectric constant of a vacuum.

Meanwhile, as shown in Figure S4C, with the increase of working distance,  $V_{oc}$  decreased from ~5.0 to ~0.4 V. This maybe because at a fixed frequency the speed of moving the triboelectric layer decreased with the increase of the working distance, resulting in a slower electron transfer rate and a lower current output. In addition, the device showed superior stability, and it could run stably for 10<sup>6</sup> cycles (Figure S4D–F).

**3.4. BS-TENG was Applied for Mechanical Energy Harvesting.** The BS-TENG is expected to be used for biomechanical energy harvesting due to its good electrical output performance. In order to investigate the effective output performance of the BS-TENG, we measured the  $V_{oc}$  and  $I_{sc}$  of the BS-TENG under different external load resistances (100 k $\Omega$  to 1 G $\Omega$ ). The effective power densities of the BS-TENG were calculated according to formula 2.

$$P = \frac{U^2}{RA} \tag{2}$$



**Figure 5.** Physiological signals and joint movement monitoring of BS-TENGs. (A) Abdominal respiratory monitoring by mounting the BS-TENG on the human belly. (B)  $V_{oc}$  of pronunciation of "A", "B", "C" to detect vocal cord vibrations.  $V_{oc}$  of the BS-TENG attached to the finger (C) and the wrist (D) to detect the bending angles.  $V_{oc}$  of the BS-TENG with (E) finger swiping and (F) tapping.

Where *R* is the load resistance and *A* is the effective area of the BS-TENG.

As shown in the blue line in Figure 4A, the  $V_{\rm oc}$  increased with the increase of the external load resistance, while the  $I_{\rm sc}$  decreased according to Ohm's law (the green line). The instantaneous power density showed an upward trend at 4 M $\Omega$ , reaching a maximum power density of about 23.2 mW m<sup>-2</sup>, and then decreased (Figure 4B).

The TENG can power electronics by harvesting mechanical energy and converting it into electricity. However, because it produces an alternating current, the collected energy needs to be rectified and stored for later use. To verify the performance of BS-TENGs, we integrated the TENG, bridge rectifier, commercial capacitors, and electronic components to construct a self-powered system, as shown in Figure 4C. The BS-TENG showed different charging capacity for different capacitors. As shown in Figure 4D, within 1 Hz and 40 s, a 1  $\mu$ F capacitor could be charged to 3.1 V, while a 2.2  $\mu$ F capacitor can only be charged to 1.8 V. With the increase of capacitance, the charging speed gradually slowed and the charging time was prolonged. In addition, commercial capacitors were charged at different application frequencies (1-5 Hz), and the corresponding voltage changes were recorded to evaluate their charging capacity, as shown in Figure 4E, where the charging speed gradually increased with the increase in frequencies. The 10  $\mu$ F capacitor could be charged to 2.8 V in 100 s at 5.0 Hz. When the switch was turned on, the electricity stored by the TENG can light up a led with a rated voltage of 2.8 V. The

above results indicated that the TENG can be used as a selfpowered source to drive low-power electronics.

**3.5.** Applications of BS-TENGs in Self-Powered Sensing. The sensing properties, such as response hysteresis, response time, strain response sensitivity, and response range to pressure, were key parameters of flexible sensors. The relationship between  $V_{oc}$  and external force was discussed. Oscilloscope and Mark-10 test system were used to collect multiple electrical signals and force signals. When the load was 11.2 N, the working distance was 3 mm, both the rising and falling speeds of the linear motor were 50 mm/min, the maximum  $V_{oc}$  was 5.5 V, the response hysteresis ( $\Delta t_1$ ) was 0.286 s, and the response time ( $\Delta t_2$ ) was 2.5 s (Figure SSA). Within the pressure range of 0 to 15 N, the relationship between the output voltage and force of BS-TENG could be satisfied by formula 3:

$$V_{\rm oc}(F) = -0.0455 + 0.4948F \tag{3}$$

The experimental data agreed well with the calculated equations. As shown in Figure S5B, the linearity  $R^2$  was up to 99.59%, and the sensitivity could reach 49.48 mV/N.

Moreover, the BS-TENG could detect 1 kHz vibration signals from a small loudspeaker and convert them to electrical signals in real time. According to the results in Figure S5C, the response time was as fast as 0.5 ms. The results further proved that the BS-TENG has a good sensing performance.

For the elastic BS-TENGs, the sensing properties, tensile properties, and output under different tensile strains were also necessary for their wide applications. To ensure the accuracy of

www.acsami.org



**Figure 6.** In vitro degradation of BS-TENG in 1× PBS buffer in a cell-culture dish (inner diameter: 56 mm) at room temperature (25 °C) for 90 days, 37 °C for 60 days, and room temperature (25 °C) with lipase for 45 days.

the results, the test speed used in this work was fixed at 1 m s<sup>-1</sup>. Since the induced strain on the skin surface and joint motion does not exceed 60% strain, the test ranges from original length to 60% strain (Figure S4E). As shown in Figure S4B–D, the output of BS-TENGs increased with the increase of strain. When 60% strain occurred, the  $V_{oct} I_{sct}$  and  $Q_{sc}$  of the device were increased by 6, 9, and 5 times, respectively.

The BS-TENG had good flexibility and stretchability, and the device could generate different electrical signals under mechanical deformation. Therefore, it can be accurately attached to the skin of neck, wrist, and fingers as a biomechanical sensor for human activity monitoring.

Breathing is one of the important vital signs to assess the body and sleep state. A complete respiratory signal consists of peaks and troughs caused by inspirations and expirations. The as-fabricated BS-TENG was encapsulated by poly(lactic acid) (PLA) due to its biocompatibility and biosafety.<sup>19,42,43</sup> For the encapsulation, the BS-TENG was placed in the middle of two PLA membranes with dimensions of 25 mm  $\times$  15 mm  $\times$  0.1 mm, and the four sides of PLA films were sealed by a heatsealing machine to eliminate any gaps. The encapsulation layer could ensure that the BS-TENG is protected from external environment, such as sweat. When the TENG was attached to the human abdomen, repeated signals under breathing were recorded and the amplitude of the voltage signal was used to detect and distinguish shallow and deep breaths (Figure 5A). To further demonstrate its good biomechanical sensing performance, the TENG was placed in the throat to monitor vocal fold vibration. When "A", "B", and "C" were emitted, 1-2.5 mV voltage signals were obtained (Figure 5B). After analyzing the signals, different letter sounds can be clearly identified.

In addition, the TENG can also be used to monitor other human motion signals. The sensor was placed on the joint. When bending 30°, 45°, 60°, and 90°, the sensor could effectively detect a distinct output signal in the form of a voltage (Figure 5C). Therefore, by analyzing the peak of  $V_{\rm OC}$ , we can accurately track the angle at which the finger was bent. As shown in Figure 5D, the device can also be used to monitor wrist movements. When the wrist was bent back and forth, a continuous voltage signal was detected. The sensors can also be provided to monitor varying amounts of forces applied by the finger under tapping or sliding. With the output variation, the degree of tapping (Figure 5E) or sliding (Figure 5F) of the finger on the device can be continuously sensed. In conclusion, PEGDA/Lap is an excellent material for the manufacture of self-powered sensors to detect human motion, including body movement and physiological signals.

3.6. Study on the Regulation of Degradability. To further investigate the degradability of the prepared BS-TENG, it was prepared with a size of 10 mm  $\times$  10 mm  $\times$  1 mm and placed in 10 mL of 1× PBS buffer for 90 days at room temperature. The degradation of the hydrogel was monitored in real time during the degradation process (Figure 6). The fresh PBS buffer solution was changed daily to ensure the same chemical environment. It can be observed that the device rapidly swelled within 3 days, and the cracks appeared on the 30th day. It was disintegrated on the 60th day and completely disappeared on the 90th day. During the degradation, the output performance of BS-TENGs in 1× PBS buffer at room temperature was studied. As shown in Figure S6, the device can operate stably for several days at first. With the swelling of encapsulation materials, the output decreased. After 7 days, it was followed by device failure, which may be attributed to the destruction of the encapsulation layer. In addition, to regulate the degradation time of the BS-TENG, it was placed in 10 mL of 1× PBS buffer for 60 days at 37 °C. The degradation cycle was shortened obviously. The device swelled rapidly within 1 day, showed obvious fracture traces on the 21st day, decomposed into fragments on the 45th day, and disappeared completely after 60 days. In addition, the degradation behavior of the device under lipase was also studied. The results showed that the presence of lipase does not affect the initial swelling behavior of the device and subsequent device damage. With the extension of time, the fragmentation of the device was accelerated, and the volume of the device decreased rapidly and disappeared completely within 45 days. Therefore, the controlled degradation of BS-TENGs can be achieved by changing the temperature or by introducing lipase into the system.

# **ACS Applied Materials & Interfaces**

## 4. DISCUSSION

Due to the choice of materials, BS-TENG had good durability. The device relied on contact and separation of the materials to generate electrical signals. The pressure of the device was relatively average, and it was not easy to damage under a large strain. However, the output of piezoelectric devices depended on the degree of bending, and the local stress was large, which could easily lead to fracture (Table S1). Therefore, the BS-TENG has advantages in terms of sensitivity and durability compatibility.

Improving the linearity and measuring range of the triboelectric sensor is a challenge at present. The linearity of some devices was limited, and it was difficult to meet the requirements of biological pressure measurement. In order to improve the measuring range and linearity, PEGDA/Laponite composites were selected to achieve performance improvement.

For chemically cross-linked hydrogels, the molecular weight  $(M_{\rm c})$  of cross-linked molecules plays an important role in determining their overall mechanical strength. Hydrogel networks with larger M<sub>c</sub> usually exhibit enhanced toughness. As the  $M_c$  increases, the stretching of polymer chains in the hydrogel network will reduce. At the same time, due to the low cross-linking density, the hydrogels with large  $M_c$  tend to exhibit a lower compression modulus. Thus, there is a trade-off between modulus and toughness in traditional chemically cross-linked hydrogel networks. In this study, the properties of PEGDA/Lap composite hydrogels were investigated. The results showed that compared with conventional hydrogels, the compression and tensile properties of PEGDA-Lap hydrogels were significantly improved with the increase of Laponite nanoparticles from 0 to 10 wt %. The enhanced mechanical properties may be attributed to the energy dissipation effect of the rigid Laponite nanoparticles in the hydrogel and the secondary interaction between the Laponite nanoparticles and PEGDA chains. However, when the content of Lap increased to 20 wt %, the mechanical properties of PEGDA/Lap hydrogels decreased slightly, which may be attributed to the limited interaction sites on the hydrogel with Laponite nanoparticles and the weakened effects between PEGDA and Laponite.

In addition, the degradability was the main feature of the devices. The degradation rate of BS-TENGs could be effectively controlled by adjusting the temperature and introducing enzymes.

Based on the PEGDA/Lap hydrogel, a BS-TENG was constructed, and its degradability was investigated. By comparing the degradation performance of devices at different ambient temperatures, we found that the temperature can accelerate the degradation. This may be since the degradation process of PEGDA/Lap hydrogel is roughly divided into three stages: swelling, cracking, and degradation. In the swelling stage, as the temperature rises, water molecules could enter the hydrogel faster, making the hydrogel quickly reach swelling equilibrium. With the increase of temperature, the secondary interaction between Laponite nanoparticles and PEGDA chains became weaker, and the long chain of PEGDA oligomer changed to a short chain faster, and the material cracked. When the material was degraded, the percentage of activated molecules increased with the temperature increase, the effective collision between molecules was enlarged, the

www.acsami.org

chemical reaction rate was accelerated, and the material would disappear in the solution faster.

Esterase is a general term for a class of enzymes with the ability to hydrolyze ester bonds. It is widely found in animals, plants, and microorganisms and can participate in a variety of biochemical reactions. As shown in eq 4, esters can be cut into acids and alcohols through hydrolysis with the participation of water molecules.

R-O-R' + H-OH----R-H + R'-OH (4)

where R-O-R' is an ester, R-H is an organic or inorganic acid, and R'-OH is a mono or polyol, aliphatic, or aromatic alcohol.

In this experiment, lipase was selected as a representative to study its regulatory effect on the degradation of BS-TENGs. Based on the existing experimental results, we guessed that in the presence of water, esterase can only destroy the fracture of ester bond in PEGDA/Lap composite hydrogels, making the material rapidly vary from long chain to short chain and finally completely degrade. Specifically, with the participation of water molecules, through hydrolysis, lipase could cut esters into acids and alcohols and then gradually decompose into water, carbon dioxide, etc. The whole process is biosafe and does not produce any harmful substances. Therefore, esterase can be selected to regulate the degradation of BS-TENGs.

In conclusion, we prepared BS-TENGs based on a stretchable conductive PEGDA/Lap hydrogel by regulating the doping of Laponite. The hydrogel has good mechanical properties, conductivity, and biocompatibility. The BS-TENGs based on this material can convert biomechanical energy into electricity, which can be used not only to power electronics but also to monitor physiological signals. Comparing with some previous works,<sup>44–49</sup> the BS-TENGs showed great stability, biodegradability, stretchability, and sensitivity (Table S1). Our data, along with the growing emergence of other studies, will demonstrate the potential applications of such nanocomposite hydrogels with enhanced mechanical properties in tissue engineering 3D scaffolds, biosensors, and transient electronics.

## 5. CONCLUSIONS

By optimizing the ratios of PEGDA and Laponite, we developed a stretchable, conductive, and biocompatible PEGDA/Lap composite hydrogel. The biodegradable BS-TENGs constructed on this basis can convert biomechanical energy into electricity and provide the monitoring of physiological signals. The strain of PEGDA/Lap composite hydrogel was about 1001.8%, and the resistance was about 10.8  $\Omega.$  Based on this, the BS-TENG with an effective area of 20 mm  $\times$  10 mm produced an open-circuit voltage of 10.4 V in single-electrode mode and could work stably for 10<sup>6</sup> cycles. The device can not only provide continuous electricity to commercial electronics by collecting biomechanical energy but also act as a sensor to monitor abdominal breathing and different human motions in real-time. In addition, the device had a controlled degradation performance and fully degraded within 90 days in  $1 \times PBS$  buffer solution. By regulation of the temperature of the degradation environment or the introduction of lipase, the device could disappear rapidly. Therefore, the BS-TENGs can be applied as self-powered electronics in the biomedical field.

## ASSOCIATED CONTENT

#### Data Availability Statement

Data are contained within article and Supporting Information.

## **Supporting Information**

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsami.2c22359.

Figure S1, characterization of PEGDA/Lap hydrogels; Figure S2, conductivity and mechanical properties of PEGDA hydrogels with different Laponite doping; Figure S3, attachment, proliferation, and morphology of the L929 cells at different times; Figure S4, electrical properties of BS-TENGs; Figure S5, research about the relationship between the output voltage of BS-TENGs and the applied force; Figure S6, output of the BS-TENG at several time intervals; Table S1, comparison of the bioresorbable self-powered mechanical sensor (PDF)

# AUTHOR INFORMATION

#### Corresponding Authors

Zhou Li – Beijing Institute of Nanoenergy and Nanosystems, Chinese Academy of Sciences, Beijing 101400, China; School of Nanoscience and Technology, University of Chinese Academy of Sciences, Beijing 100049, P. R. China;
orcid.org/0000-0002-9952-7296; Email: zli@ binn.cas.cn

Yulin Deng – School of Life, Beijing Institute of Technology, Beijing 100081, China; <sup>●</sup> orcid.org/0000-0002-0720-4052; Email: deng@bit.edu.cn

#### Authors

- Zhe Li School of Medical Technology, Institute of Engineering Medicine, Beijing Institute of Technology, Beijing 100081, China
- **Cong Li** Beijing Institute of Nanoenergy and Nanosystems, Chinese Academy of Sciences, Beijing 101400, China
- Wei Sun Beijing Institute of Nanoenergy and Nanosystems, Chinese Academy of Sciences, Beijing 101400, China; School of Nanoscience and Technology, University of Chinese Academy of Sciences, Beijing 100049, P. R. China
- **Yuan Bai** Beijing Institute of Nanoenergy and Nanosystems, Chinese Academy of Sciences, Beijing 101400, China

Complete contact information is available at: https://pubs.acs.org/10.1021/acsami.2c22359

## **Author Contributions**

Zhe Li designed the research. Zhe Li and C.L. collected the data. Zhe Li and Y.B. processed the data. W.S. carried out COMSOL simulation. Zhe Li wrote the original manuscript. Zhou Li and Y.L.D. provided suggestions and improved the manuscript. Zhe Li and Zhou Li obtained funding. Y.L.D. provides resources. All authors have read and agreed to publish the manuscript.

#### Funding

This research was funded by the Beijing Natural Science Foundation (7224368, JQ20038, L212010), the National Natural Science Foundation of China (T2125003, 61875015, 82202348), the Strategic Priority Research Program of the Chinese Academy of Sciences (XDA16021101), and the China Postdoctoral Science Foundation (2022T150052).

#### Notes

The authors declare no competing financial interest.

# ACKNOWLEDGMENTS

The authors thank Zhao Li, Jian Liu, and Fangzhou Li for their assistance in perfecting this article.

# REFERENCES

(1) Chu, S.; Majumdar, A. Opportunities and Challenges for A Sustainable Energy Future. *Nature* **2012**, *488* (7411), 294–303.

(2) Song, Y. D.; Wang, N.; Hu, C. S.; Wang, Z. L.; Yang, Y. Soft Triboelectric Nanogenerators for Mechanical Energy Scavenging and Self-Powered Sensors. *Nano Energy* **2021**, *84*, 105919.

(3) Yu, D. J.; Li, Z.; Xie, W. Q.; Li, D. S.; Li, Z.; Li, Y. S. Applications of Nanogenerator-Based Wearable Devices in Orthopedics. *Nano Energy* **2022**, *103*, 107762.

(4) Sun, M. J.; Li, Z.; Yang, C. Y.; Lv, Y. J.; Yuan, L.; Shang, C. X.; Liang, S. Y.; Guo, B. W.; Liu, Y.; Li, Z.; Luo, D. Nanogenerator-Based Devices for Biomedical Applications. *Nano Energy* 2021, 89, 106461.
(5) He, T. Y. Y.; Wang, H.; Wang, J. H.; Tian, X.; Wen, F.; Shi, Q.
F.; Ho, J. S.; Lee, C. K. Self-Sustainable Wearable Textile Nano-Energy Nano-System (NENS) for Next-Generation Healthcare Applications. *Adv. Sci.* 2019, 6 (24), 1901437.

(6) Lee, J.; Zambrano, B. L.; Woo, J.; Yoon, K.; Lee, T. Recent Advances in 1D Stretchable Electrodes and Devices for Textile and Wearable Electronics: Materials, Fabrications, and Applications. *Adv. Mater.* **2020**, 32 (5), e1902532.

(7) Chung, H. U.; Kim, B. H.; Lee, J. Y.; Lee, J.; Xie, Z. Q.; Ibler, E. M.; Lee, K.; Banks, A.; Jeong, J. Y.; Kim, J.; et al. Binodal, Wireless Epidermal Electronic Systems with in-Sensor Analytics for Neonatal Intensive Care. *Science* **2019**, *363* (6430), 947.

(8) Li, Y. Y.; Zhang, Y. H.; Yi, J.; Peng, X.; Cheng, R. W.; Ning, C.; Sheng, F. F.; Wang, S.; Dong, K.; Wang, Z. L. Large-scale Fabrication of Core-Shell Triboelectric Braided Fibers and Power Textiles for Energy Harvesting and Plantar Pressure Monitoring. *Ecomat* **2022**, *4* (4), e12191.

(9) Zhang, C.; Guo, Z. H.; Zheng, X. X.; Zhao, X. J.; Wang, H. L.; Liang, F.; Guan, S.; Wang, Y.; Zhao, Y. B.; Chen, A. H.; Zhu, G.; Wang, Z. L. A Contact-Sliding-Triboelectrification-Driven Dynamic Optical Transmittance Modulator for Self-Powered Information Covering and Selective Visualization. *Adv. Mater.* **2020**, *32* (1), 1904988.

(10) Fu, W. B.; Turcheniuk, K.; Naumov, O.; Mysyk, R.; Wang, F. J.; Liu, M.; Kim, D.; Ren, X. L.; Magasinski, A.; Yu, M. H.; Feng, X. L.; Wang, Z. L.; Yushin, G. Materials and Technologies for Multifunctional, Flexible or Integrated Supercapacitors and Batteries. *Mater. Today* **2021**, *48*, 176–197.

(11) Li, Y. Y.; Wei, C. H.; Jiang, Y.; Cheng, R. W.; Zhang, Y. H.; Ning, C.; Dong, K.; Wang, Z. L. Continuous Preparation of Chitosan-Based Self-Powered Sensing Fibers Recycled from Wasted Materials for Smart Home Applications. *Adv. Fiber Mater.* **2022**, *4*, 1584.

(12) Xu, W.; Huang, L. B.; Wong, M. C.; Chen, L.; Bai, G. X.; Hao, J. H. Environmentally Friendly Hydrogel-Based Triboelectric Nanogenerators for Versatile Energy Harvesting and Self-Powered Sensors. *Adv. Energy Mater.* **2017**, *7* (1), 1601529.

(13) Li, H.; Chang, T. R.; Gai, Y. S.; Liang, K.; Jiao, Y. L.; Li, D. F.; Jiang, X. R.; Wang, Y.; Huang, X. C.; Wu, H.; Liu, Y. M.; Li, J.; Bai, Y. M.; Geng, K.; Zhang, N. A. R.; Meng, H.; Huang, D. S.; Li, Z.; Yu, X. G.; Chang, L. Q. Human Joint Enabled Flexible Self-sustainable Sweat Sensors. *Nano Energy* **2022**, *92*, 106786.

(14) Qu, X. C.; Liu, Z.; Tan, P. C.; Wang, C.; Liu, Y.; Feng, H. Q.; Luo, D.; Li, Z.; Wang, Z. L. Artificial Tactile Perception Smart Finger for Material Identification Based on Triboelectric Sensing. *Science Advances* **2022**, *8* (31), eabq2521.

(15) Cheng, R. W.; Dong, K.; Chen, P. F.; Ning, C.; Peng, X.; Zhang, Y. H.; Liu, D.; Wang, Z. L. High Output Direct-Current Power Fabrics Based on the Air Breakdown Effect. *Energy Environ. Sci.* **2021**, *14* (4), 2460–2471.

(16) Dong, K.; Peng, X.; An, J.; Wang, A. C.; Luo, J. J.; Sun, B. Z.; Wang, J.; Wang, Z. L. Shape Adaptable and Highly Resilient 3D Braided Triboelectric Nanogenerators as e-Textiles for Power and Sensing. *Nat. Commun.* **2020**, *11* (1), 2868.

(17) Zheng, Q.; Zou, Y.; Zhang, Y.; Liu, Z.; Shi, B.; Wang, X.; Jin, Y.; Ouyang, H.; Li, Z.; Wang, Z. L. Biodegradable Triboelectric Nanogenerator as A Life-Time Designed Implantable Power Source. *Sci. Adv.* **2016**, *2* (3), e1501478.

(18) Sun, J.; Guo, H.; Ribera, J.; Wu, C.; Tu, K.; Binelli, M.; Panzarasa, G.; Schwarze, F.; Wang, Z. L.; Burgert, I. Sustainable and Biodegradable Wood Sponge Piezoelectric Nanogenerator for Sensing and Energy Harvesting Applications. *ACS Nano* **2020**, *14* (11), 14665–14674.

(19) Li, Z.; Feng, H. Q.; Zheng, Q.; Li, H.; Zhao, C. C.; Ouyang, H.; Noreen, S.; Yu, M.; Su, F.; Liu, R. P.; Li, L. L.; Wang, Z. L.; Li, Z. Photothermally Tunable Biodegradation of Implantable Triboelectric Nanogenerators for Tissue Repairing. *Nano Energy* **2018**, *54*, 390– 399.

(20) Wang, H. L.; Guo, Z. H.; Zhu, G.; Pu, X.; Wang, Z. L. Boosting the Power and Lowering the Impedance of Triboelectric Nanogenerators through Manipulating the Permittivity for Wearable Energy Harvesting. *ACS Nano* **2021**, *15* (4), 7513–7521.

(21) Ning, C.; Cheng, R. W.; Jiang, Y.; Sheng, F. F.; Yi, J.; Shen, S.; Zhang, Y. H.; Peng, X.; Dong, K.; Wang, Z. L. Helical Fiber Strain Sensors Based on Triboelectric Nanogenerators for Self-Powered Human Respiratory Monitoring. *ACS Nano* **2022**, *16* (2), 2811–2821.

(22) Chen, X. X.; Song, Y.; Chen, H. T.; Zhang, J. X.; Zhang, H. X. An Ultrathin stretchable Triboelectric Nanogenerator with Coplanar Electrode for Energy Harvesting and Gesture Sensing. *Journal of Materials Chemistry A* **2017**, 5 (24), 12361–12368.

(23) Bao, D. Q.; Wen, Z.; Shi, J. H.; Xie, L. J.; Jiang, H. X.; Jiang, J. X.; Yang, Y. Q.; Liao, W. Q.; Sun, X. H. An Anti-Freezing Hydrogel Based Stretchable Triboelectric Nanogenerator for Biomechanical Energy Harvesting at Sub-Zero Temperature. *Journal of Materials Chemistry A* **2020**, *8* (27), 13787–13794.

(24) Xu, W.; Huang, L. B.; Hao, J. H. Fully Self-Healing and Shape-Tailorable Triboelectric Nanogenerators Based on Healable Polymer and Magnetic-Assisted Electrode. *Nano Energy* **201**7, *40*, 399–407.

(25) Yang, C. H.; Suo, Z. G. Hydrogel Ionotronics. Nat. Rev. Mater. 2018, 3 (6), 125–142.

(26) Keplinger, C.; Sun, J. Y.; Foo, C. C.; Rothemund, P.; Whitesides, G. M.; Suo, Z. G. Stretchable, Transparent, Ionic Conductors. *Science* **2013**, *341* (6149), 984–987.

(27) Lu, B. Y.; Yuk, H.; Lin, S. T.; Jian, N. N.; Qu, K.; Xu, J. K.; Zhao, X. H. Pure PEDOT:PSS Hydrogels. *Nat. Commun.* **2019**, *10* (1), 1043.

(28) Yuk, H.; Lu, B. Y.; Zhao, X. H. Hydrogel Bioelectronics. *Chem.* Soc. Rev. 2019, 48 (6), 1642–1667.

(29) Yuan, Y. X.; Zhou, J. L.; Lu, G. Q.; Sun, J. X.; Tang, L. Q. Highly Stretchable, Transparent, and Self-Adhesive Ionic Conductor for High-Performance Flexible Sensors. *Acs Appl. Polym. Mater.* **2021**, 3 (3), 1610–1617.

(30) Qu, X. Y.; Wang, S. Y.; Zhao, Y.; Huang, H.; Wang, Q.; Shao, J. J.; Wang, W. J.; Dong, X. C. Skin-Inspired Highly Stretchable, Tough and Adhesive Hydrogels for Tissue-Attached Sensor. *Chem. Eng. J.* **2021**, *425*, 131523.

(31) Huang, R. K.; Zhang, X. X.; Li, W. Z.; Shang, L. R.; Wang, H.; Zhao, Y. J. Suction Cups-Inspired Adhesive Patch with Tailorable Patterns for Versatile Wound Healing. *Adv. Sci.* **2021**, *8* (17), e2100201.

(32) Chen, X. B.; Zhang, J. L.; Wu, K. T.; Wu, X. H.; Tang, J. Y.; Cui, S. Q.; Cao, D. L.; Liu, R. L.; Peng, C.; Yu, L.; Ding, J. D. Visualizing the In Vivo Evolution of An Injectable and Thermosensitive Hydrogel Using Tri-Modal Bioimaging. *Small Methods* **2020**, *4* (9), 2000310.

(33) Wang, C.; Hu, K.; Zhao, C. C.; Zou, Y.; Liu, Y.; Qu, X. C.; Jiang, D. J.; Li, Z.; Zhang, M. R.; Li, Z. Customization of Conductive (36) Chang, C. W.; van Spreeuwel, A.; Zhang, C.; Varghese, S. PEG/ clay Nnanocomposite Hydrogel: A Mechanically Robust Tissue Engineering Scaffold. *Soft Matter* **2010**, *6* (20), 5157–5164.

Elastomer Based on PVA/PEI for Stretchable Sensors. Small 2020, 16

(34) Baghdadi, H. A.; Sardinha, H.; Bhatia, S. R. Rheology and

Gelation Kinetics in Laponite Dispersions Containing Poly(Ethylene

(35) Wu, Y. H.; Yao, Y.; Zhang, J. M.; Gui, H.; Liu, J. J.; Liu, J. F.

Tumor-Targeted Injectable Double-Network Hydrogel for Prevention

of Breast Cancer Recurrence and Wound Infection via Synergistic

Oxide). J. Polym. Sci. Pol Phys. 2005, 43 (2), 233-240.

(37) Niu, S. M.; Wang, S. H.; Lin, L.; Liu, Y.; Zhou, Y. S.; Hu, Y. F.; Wang, Z. L. Theoretical Study of Contact-mode Triboelectric Nanogenerators as An Affective Power Source. *Energy Environ. Sci.* **2013**, 6 (12), 3576–3583.

(38) Shin, S. H.; Bae, Y. E.; Moon, H. K.; Kim, J.; Choi, S. H.; Kim, Y.; Yoon, H. J.; Lee, M. H.; Nah, J. Formation of Triboelectric Series via Atomic-Level Surface Functionalization for Triboelectric Energy Harvesting. *ACS Nano* **2017**, *11* (6), 6131–6138.

(39) Niu, S. M.; Wang, Z. L. Theoretical Systems of Triboelectric Nanogenerators. *Nano Energy* **2015**, *14*, 161–192.

(40) Dong, K.; Deng, J. N.; Zi, Y. L.; Wang, Y. C.; Xu, C.; Zou, H. Y.; Ding, W. B.; Dai, Y. J.; Gu, B. H.; Sun, B. Z.; Wang, Z. L. 3D Orthogonal Woven Triboelectric Nanogenerator for Effective Biomechanical Energy Harvesting and as Self-Powered Active Motion Sensors. *Adv. Mater.* **2017**, *29* (38), 1702648.

(41) Liu, R. Y.; Kuang, X.; Deng, J. N.; Wang, Y. C.; Wang, A. C.; Ding, W. B.; Lai, Y. C.; Chen, J.; Wang, P. H.; Lin, Z. Q.; Qi, H. J.; Sun, B. Q.; Wang, Z. L. Shape Memory Polymers for Body Motion Energy Harvesting and Self-Powered Mechanosensing. *Adv. Mater.* **2018**, 30 (8), 1705195.

(42) Fairag, R.; Rosenzweig, D. H.; Ramirez-Garcialuna, J. L.; Weber, M. H.; Haglund, L. Three-Dimensional Printed Polylactic Acid Scaffolds Promote Bone-Like Matrix Deposition in Vitro. *ACS Appl. Mater. Interfaces* **2019**, *11* (17), 15306–15315.

(43) Cifuentes, S. C.; Gavilan, R.; Lieblich, M.; Benavente, R.; Gonzalez-Carrasco, J. L. In Vitro Degradation of Biodegradable Polylactic Acid/Magnesium Composites: Relevance of Mg Particle Shape. *Acta Biomaterialia* **2016**, *32*, 348–357.

(44) Ouyang, H.; Li, Z.; Gu, M.; Hu, Y. R.; Xu, L. L.; Jiang, D. J.; Cheng, S. J.; Zou, Y.; Deng, Y.; Shi, B. J.; Hua, W.; Fan, Y. B.; Li, Z.; Wang, Z. L. A Bioresorbable Dynamic Pressure Sensor for Cardiovascular Postoperative Care. *Adv. Mater.* **2021**, *33* (39), 2102302.

(45) Curry, E. J.; Le, T. T.; Das, R.; Ke, K.; Santorella, E. M.; Paul, D.; Chorsi, M. T.; Tran, K. T. M.; Baroody, J.; Borges, E. R.; Ko, B.; Golabchi, A.; Xin, X. N.; Rowe, D.; Yue, L. X.; Feng, J. L.; Morales-Acosta, M. D.; Wu, Q.; Chen, I. P.; Cui, X. T.; Pachter, J.; Nguyen, T. D. Biodegradable Nanofiber-Based Piezoelectric Transducer. *Proc. Natl. Acad. Sci. U.S.A.* **2020**, *117* (1), 214–220.

(46) Luo, Y. C.; Yu, M. L.; Zhang, Y. T.; Wang, Y. Y.; Long, L.; Tan, H. H.; Li, N.; Xu, L. J.; Xu, J. X. Highly Sensitive Strain Sensor and Self-Powered Triboelectric Nanogenerator Using A Fully Physical Crosslinked Double-Network Conductive Hydrogel. *Nano Energy* **2022**, *104*, 107955.

(47) Zhang, W. Y.; Liu, Q.; Chao, S. Y.; Liu, R. P.; Cui, X.; Sun, Y.; Ouyang, H.; Li, Z. Ultrathin Stretchable Triboelectric Nanogenerators Improved by Postcharging Electrode Material. *ACS Appl. Mater. Interfaces* **2021**, *13* (36), 42966–42976.

(48) Dai, X. H.; Long, Y.; Jiang, B.; Guo, W. B.; Sha, W.; Wang, J. W.; Cong, Z. F.; Chen, J. W.; Wang, B. J.; Hu, W. G. Ultra-Antifreeze, Ultra-Stretchable, Transparent, and Conductive Hydrogel for Multi-Functional Flexible Electronics as Strain Sensor and Triboelectric Nanogenerator. *Nano Research* **2022**, *15* (6), 5461–5468.

(49) Long, Y.; Wang, Z.; Xu, F.; Jiang, B.; Xiao, J. F.; Yang, J.; Wang, Z. L.; Hu, W. G. Mechanically Ultra-Robust, Elastic, Conductive, and Multifunctional Hybrid Hydrogel for A Triboelectric Nanogenerator and Flexible/Wearable Sensor. *Small* **2022**, *18* (47), 2203956.

(7), e1904758.