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Robust and durable triboelectric nanogenerators enabled by a mechanically strong and mildly healable polymer†

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The capability of healing structural damage is highly desired for triboelectric nanogenerators (TENGs), which are subject to repeated mechanical loading. However, it remains difficult to balance the intrinsic mechanical strength and healing capability of the materials used in TENGs. Herein, we exploit a mechanically strong and mildly healable polymer (THP) as the key material of TENGs. THP is based on polyurethane–urea containing coordinate bonds, and its healing can be triggered by simply wetting the breakage area with water or alcoholic drinks. The healing ratio and the tensile strength of THP can reach ~90% and ~11.6 MPa, respectively. Correspondingly, the prepared THP-based TENGs (THP-TENGs) exhibit high enough robustness to withstand mechanical loading as well as excellent healability for recovering device functionalities after mechanical damage. In addition, disused devices can be recycled to produce new ones based on the solution processibility of THP. The fabricated THP-TENG could serve as a mechanical energy harvester as well as a self-powered sensor for detecting tactile and electrophysiological signals, suggesting utilities in advanced wearable systems.

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Introduction

The rapid development of wearable electronics and the internet of things imposes high requirements on power sources and sensors.[1–3] The triboelectric nanogenerator (TENG), a device based on the triboelectric effect and electrostatic induction, possesses excellent ability to convert ambient mechanical energy into electrical output in a miniaturized configuration. This device shows great potential as a distributed power source for emerging electronics or acting as a self-powered sensor for detecting mechanical motion.[3–7] Compared to other miniaturized energy devices, TENGs possess various advantages, including simple structure, low cost, high output voltage, and high efficiency in energy conversion for low-frequency mechanical inputs.[8–12]

During the process of energy harvesting, TENGs are subject to frequent mechanical forces for generating electrical output. This process could cause mechanical damage to the device structures, leading to reduced durability and lifespan of the system.[13,14] A promising method to solve this problem is to construct healable TENGs based on appropriate reconfigurable polymer materials.[14–16] However, there is a tradeoff between mechanical strength and healability for most healable polymers, which creates challenges for their utilization in TENGs.[17–23] For instance, autonomous-healable polymers with high molecular chain mobility can heal without an external trigger. But they usually present relatively low mechanical strength, which cannot withstand the high mechanical loading applied on TENGs.[24–29] On the other hand, devices based on non-autonomous-healable polymers, as exemplified by polymers containing Diels–Alder bonds *etc.*, could easily exhibit considerable mechanical strength at levels above 10 MPa. But their healing may require sophisticated conditions, such as high temperature, infrared radiation, or other stimuli that are not compatible with the device system.[15,19,30,31] In this regard, simplicity of the healing process aligned with the normal operating conditions of TENGs becomes essential. Despite extensive efforts, technological approaches that can achieve these attributes remain limited.

In this study, we exploit a mechanically strong and mildly healable polymer (THP) as the base material for the construction of TENGs. THP is constructed from polyurethane–urea and involves coordination interactions. The tensile strength of THP (11.6 MPa) exceeds that of most healable polymers used in TENGs, which confers excellent robustness on the devices. The healing of THP can be conducted in the presence of water or alcoholic drinks which are ubiquitous and common in daily life.

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After healing of mechanically damaged devices, the electrical output of the THP-based TENG (THP-TENG) can recover to the initial state and remain stable for 8000 operation cycles. Furthermore, based on the solvent processability of THP, disused THP-TENGs can be recycled for the production of new devices. The fabricated THP-TENG can serve as a mechanical energy harvester and a self-powered sensor for detecting tactile and electrophysiological signals simultaneously, suggesting opportunities for applications in wearable electronic systems.

Experimental section

Reagents and materials

Polytetrahydrofuran (PTMEG, $M_n = 850$), isophorone diisocyanate (IPDI), dibutyltin dilaurate (DBTDL), triethylamine (TEA), dopamine hydrochloride (DA-HCl), dimethylformamide (DMF), and calcium chloride ($CaCl_2$) were purchased from Aladdin-reagent (China). All the chemicals were used as received.

Preparation of the mechanically tough and mildly healable polymer (THP)

30 g PTMEG was heated in an oil bath of about 115 °C under vacuum and magnetic stirring for 2 h for water removal. After cooling the system to 70 °C, 16.3 g IPDI and 2 drops of DBTDL were added under mechanical stirring for sealed reaction. The temperature of the oil bath was successively raised to 105 °C for 3 h and 115 °C for 1 h to form the isocyanate terminated prepolymer (NCO-prepolymer) for further experiment. 5 g NCO-prepolymer dissolved in 5 ml DMF and 1.05 g DA-HCl dissolved in 4 ml DMF were mixed. Then 0.6 g TEA was added in two batches in 2 h followed by reaction for 24 h to get the catechol containing prepolymer (DA-NCO-prepolymer). Then, the catechol terminated prepolymer (DA-prepolymer) was obtained after the separation and precipitation in water several times during 1–2 h. The DA-prepolymer was dissolved into DMF (20 ml) by means of stirring under 90 °C. Then 0.126 g $CaCl_2$ dissolved in 5 mL DMF using a sonicator was added for mixing for 30 min. The mixture solution was transferred to a Teflon dish for the chain extension and evaporation of the solvent at 65 °C for 2 days. The resultant THP film was obtained for the fabrication of TENGs.

Preparation of the THP-TENG

The THP based TENG (THP-TENG) was fabricated by encapsulating electrodes between two THP layers. Specifically, the electrode pattern was prepared by coating Ag paste on a Teflon film through a stencil mask followed by heating and removing the solvent. Then the surface of a THP film was wetted by ethanol, and the resultant sticky THP surface can transfer the electrode pattern from the Teflon surface to the THP surface. Ni fabric tape was used to connect the electrode with the external circuit for measurement. The THP-TENG was achieved after encapsulating the electrode with another THP layer. For detecting the electromyography signals, another electrode pattern was fabricated and transferred to the bottom surface of the THP-TENG in a similar way.

Measurement of the device

The output voltage and current were characterized using a LeCroy WaveRunner oscilloscope (probe resistance value of 50 MΩ) and low noise current amplifier (Stanford Research Systems, SR570), respectively. The tensile property of the prepared polymer was measured using a universal testing machine at room temperature of about 24–27 °C and humidity of ~50%. The healing ratio was calculated based on the results of tensile curves. During the healing process, the cutting width and depth are around 0.7 cm and 0.4 mm, respectively. The amount of water added is about 100 μL. The water was added twice. The voltage–current (I–V) curves were measured using an electrochemical workstation. The electromyogram (EMG) signals were measured using a data acquisition hardware device (PowerLab26T).

Results and discussion

To achieve the expected TENG devices, the mechanically strong and mildly healable polymer (THP) was firstly prepared as the base materials of the device. As illustrated in Fig. 1, the synthesis process of THP mainly includes four steps: (1) synthesis of the isocyanate terminated prepolymer (NCO-prepolymer) by the reaction of polytetrahydrofuran (PTMEG) and isophorone diisocyanate (IPDI). (2) Synthesis of the catechol containing prepolymer (DA-NCO-prepolymer) by the reaction between the NCO-prepolymer and dopamine (the molar ratio of isocyanate to catechol ends is about 1 to 0.7, and TEA was excessive). (3) Chain extension of DA-NCO-prepolymer by the reaction between water and –NCO to form the catechol terminated prepolymer (DA-prepolymer). The structure of the product was confirmed by ^{1}H NMR (Fig. S1†) and UV-vis absorption spectroscopy (Fig. S2†). (4) Chain extension of DA-prepolymer by the coordination interaction between catechol ends and Ca^{2+}. The coordination was proved by isothermal titration calorimetry (Fig. S3†). The final THP film was formed after removing the solvent. The existence of both isocyanate and catechol ends in DA-NCO-prepolymer enables the two-step chain-extension reaction to reach the excellent comprehensive performance. The first chain extension using water generates urea groups for the improvement of mechanical strength. The second chain extension using Ca^{2+} introduces dynamic coordination bonds for further enhancing the strength of THP and endowing healability.

As the constituent material of TENGs, the mechanical properties of THP strongly affect the robustness of devices. Fig. 2a shows the tensile stress–strain curves of THP samples with different amounts of added $CaCl_2$ (C_{Ca}, which is defined as the mole percentage ratio of catechol coordinated with Ca^{2+} to the total catechol). It was found that the tensile strength of THP improves with the increase of C_{Ca}. For the sample without added $CaCl_2$, the tensile strength was about 4.8 MPa. After adding 14% $CaCl_2$ into the system, the resultant THP sample exhibits an increased tensile strength of 6.3 MPa. When the C_{Ca} enhances to 28% and 42%, the tensile strength of THP can reach 11.6 MPa and 19.7 MPa, respectively, which are higher

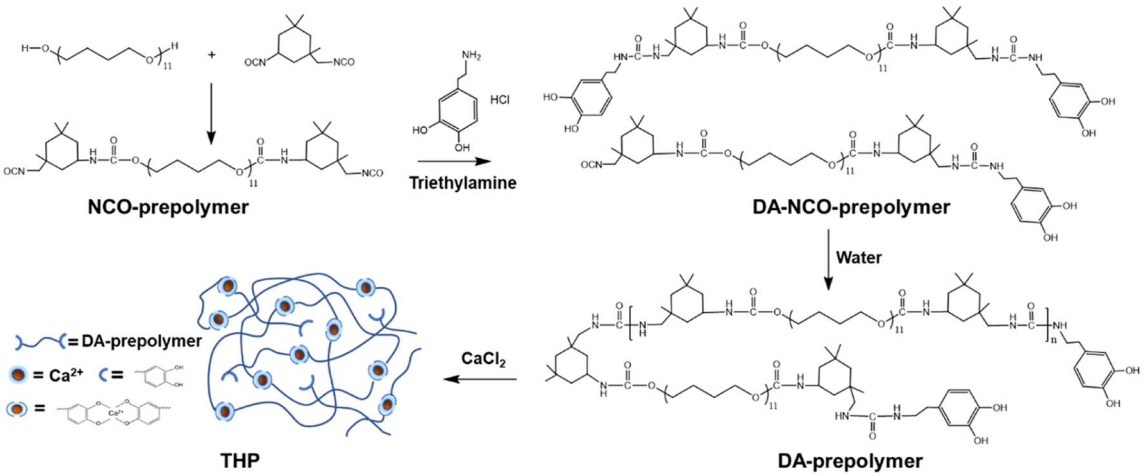

Fig. 1 The preparation process of THP.

than those of most healable polymers used in TENGs. As mentioned above, by means of the chain extension of DA-NCO-prepolymer using water, the tensile strength of the prepared DA-prepolymer can reach 4.8 MPa. The introduction of $CaCl_2$ contributes to the further chain extension of DA-prepolymer by the formation of coordination interaction, thereby leading to the declined molecular mobility and the improved strength of THP.[32,33]

The mechanical properties of THP were further evaluated by cyclic tensile tests as shown in Fig. 2b and S4.† After a strain of 50% was applied, relatively large stress–strain loops were observed. Specifically, when the strain reaches about 10%, the yield point appears, accompanied by the rupture of some intermolecular bonds and hard-segment phase. During the unloading process, a large residual strain was observed, which can be attributed to the chain entanglement and the

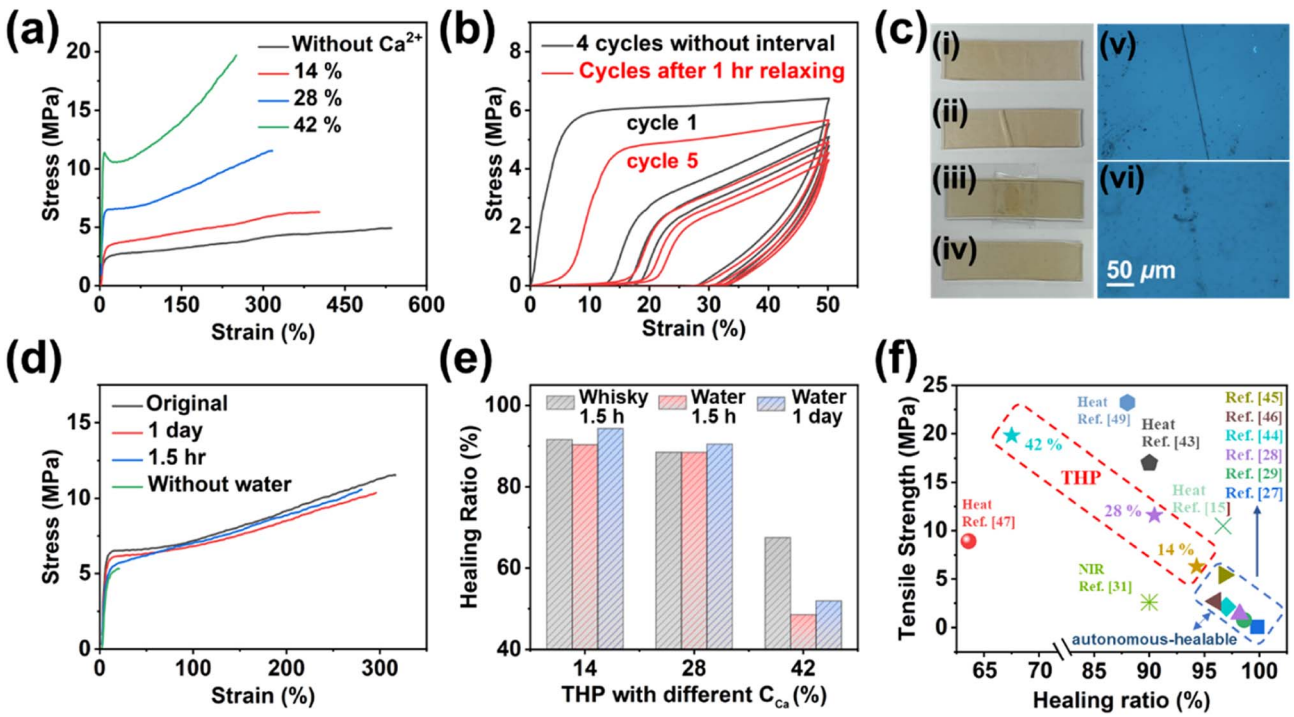

Fig. 2 The mechanical and healable properties of THP. (a) Tensile stress–strain curves of THP samples with different C_{Ca}. (b) Cyclic tensile tests of THP with C_{Ca} of 28%. (c) Photographs (i–iv) and microscope images (v and vi) illustrating the water-assisted healing process of THP. (d) The tensile stress–strain curves of THP at the original and the healed state. (e) The healing ratio of THP (C_{Ca} = 14%, 28% and 42%) under different healing treatments. (f) Comparison of the healing ratio and tensile strength of representative healable polymers employed in TENGs. Among them, the data in the blue box come from some previously reported autonomous-healable TENGs, and the data in the red box come from our THP with C_{Ca} of 14%, 28% and 42%. The others come from some previously reported non-autonomous-healable TENGs.

reconfiguration of some hydrogen bonds and coordinate bonds *etc.* After annealing the cyclically tested sample without mechanical loading for 1 h, followed by cyclic testing, the sample exhibits loops (cycle 5 and after in Fig. 2b) similar to the previous cycles, indicating the partial recovery of molecular structure driven by entropic elasticity. From the perspective of devices, it is necessary to further develop THP with low hysteresis. Some strategies, such as the optimization of the soft and hard segments, and the construction of dynamic crosslinked structure *etc.*, have been reported and deserve attempts.[34–36] In addition, the mechanical properties were also characterized by Dynamic Mechanical Analysis (DMA) as shown in Fig. S5,† where multiple transition temperatures were observed.

The as-prepared THP shows healing ability in the presence of water, and the healing process is illustrated in Fig. 2c. Specifically, the THP sample ($C_{Ca} = 28\%$) was firstly cut using a razor blade (i and ii). After re-contacting the cut ends carefully and adding a small amount of water on the breakage area, a piece of scotch tape was applied on the breakage area for retaining the wetting of this area for 1 day (iii). Then the scotch tape was removed, and the healed THP sample without a scar was achieved after the evaporation of water (iv). Optical microscope images of the breakage area show that the breakage mark (v) almost disappeared after the water-assisted healing treatment (vi), confirming the healing ability of the prepared THP.

The healability of THP can be quantitatively measured by comparing the tensile stress–strain curves of samples at the initial and the healed state, and the healing ratio is defined as the proportion of the failure strain of the healed sample relative to that of the initial one. As shown in Fig. 2d, the failure strain of the initial THP ($C_{Ca} = 28\%$) and the healed THP (after water treatment for 1 day) are 317% and 297%, respectively. Correspondingly, the healing ratio of THP was calculated to be above 90%. In contrast, the healing ratio of THP healed without application of water was only 6.6%, implying the importance of water in the healing process. The introduction of water not only enables the coordination between Ca^{2+} and catechol to become dynamic, but also plasticizes the polymer system.[37–39] Both factors lead to the increased molecular mobility of THP, promoting the diffusion of molecular chains and the structure re-establishment around breakage area for final healing.

Fig. 2e demonstrates the healing ratio of prepared THP samples with different C_{Ca} (after water treatment for 1 day). The results show that the healing ratio decreases with the increase of the C_{Ca}. When the C_{Ca} increases from 14% to 42%, the healing ratio of samples changes from 94% to 52%. As mentioned above, the introduction of $CaCl_2$ mainly contributes to the chain extension of DA-prepolymer. This means that the THP samples with higher C_{Ca} possess the larger length of molecular chains and therefore lower molecular mobility, leading to the decrease in healability. Besides water, the healing of THP can also be triggered by some alcoholic drinks, such as whisky. It was found that the sample healed by whisky shows a better healing ratio than the sample healed by water when healing time is short (1.5 h), which may be attributed to the better compatibility of alcohol with THP.

From the perspective of practical operation, the healable TENG is expected to possess not only healability for fracture recovery, but also excellent strength to resist mechanical failure. Fig. 2f and S6† summarize the mechanical strength and the healing ratio of a series of healable polymers used in TENGs.[15,19,27–29,31,40–49] It was found that our THP shows an obviously higher strength than numerous autonomous-healable polymers used in TENGs. The strength of THP is comparable to that of non-autonomous-healable polymers. However, the healing of regular non-autonomous-healable materials usually requires stimuli that are not compatible with the operation environment of TENGs, such as heat, infrared radiation *etc.* These processes are inconvenient when the devices are used in an outdoor or wearable configuration. In contrast, the prepared THP can conduct the healing easily with the assistance of water or alcoholic drinks which are ubiquitous, mild, and safe. This highlights the advantage of THP as the base material of TENGs, which balances the healing and mechanical properties. Besides, this strategy could be compatible to other polymer systems to achieve various mechanically strong and mildly healable polymers for the development of advanced healable TENGs.

Based on THP, we prepared a single-electrode-mode TENG (THP-TENG) which consists of external THP layers and an internal electrode as shown in Fig. 3a. The THP-TENG can convert mechanical energy into electricity based on the triboelectric effects, and the working principle is illustrated in Fig. 3a(iii–vi). Specifically, the contact between the paired material and the THP-TENG causes electron transfer and redistribution at the contact interface because of the difference in electron affinity (iii). When the paired material departs from the device, two oppositely charged surfaces are generated. Among them, the charged THP surface will induce a transient electron flow between the internal electrode and the ground by electrostatic effects (iv) until electrical equilibrium is achieved (v). When the charged paired material approaches the device again, the opposite electric potential and electron flow are induced (vi). By repeating this procedure driven by mechanical force, the ambient mechanical energy is successfully converted.

The paired contact material in Fig. 3a could be any material that possesses different electron affinity with THP. Herein, we take PDMS film as an example of the paired material to evaluate the electrical output performance of the THP-TENG. As shown in Fig. 3b and c, when the THP-TENG (2.5 cm × 4 cm) is driven by a linear motor with the frequency of 4 Hz, the peak output voltage and short-circuit current (I_{sc}) of the device are about 28 V and 2.86 μA, respectively. The output voltage and current change under different load resistances are shown in Fig. 3d. The peak electrical power density is calculated to be about 2.9 mW m^{-2} at a load resistance of 100 MΩ (Fig. 3e). Both the type of paired materials and the contact frequency will affect the electrical output of the device. As shown in Fig. 3f, the PDMS as the paired material could achieve a relatively high output voltage, which is ascribed to its high electron affinity compared with other materials. Meanwhile, it was found that output voltage increases with the enhancement of contact frequency from 1 Hz to 8 Hz (Fig. 3g), which is in accordance with the

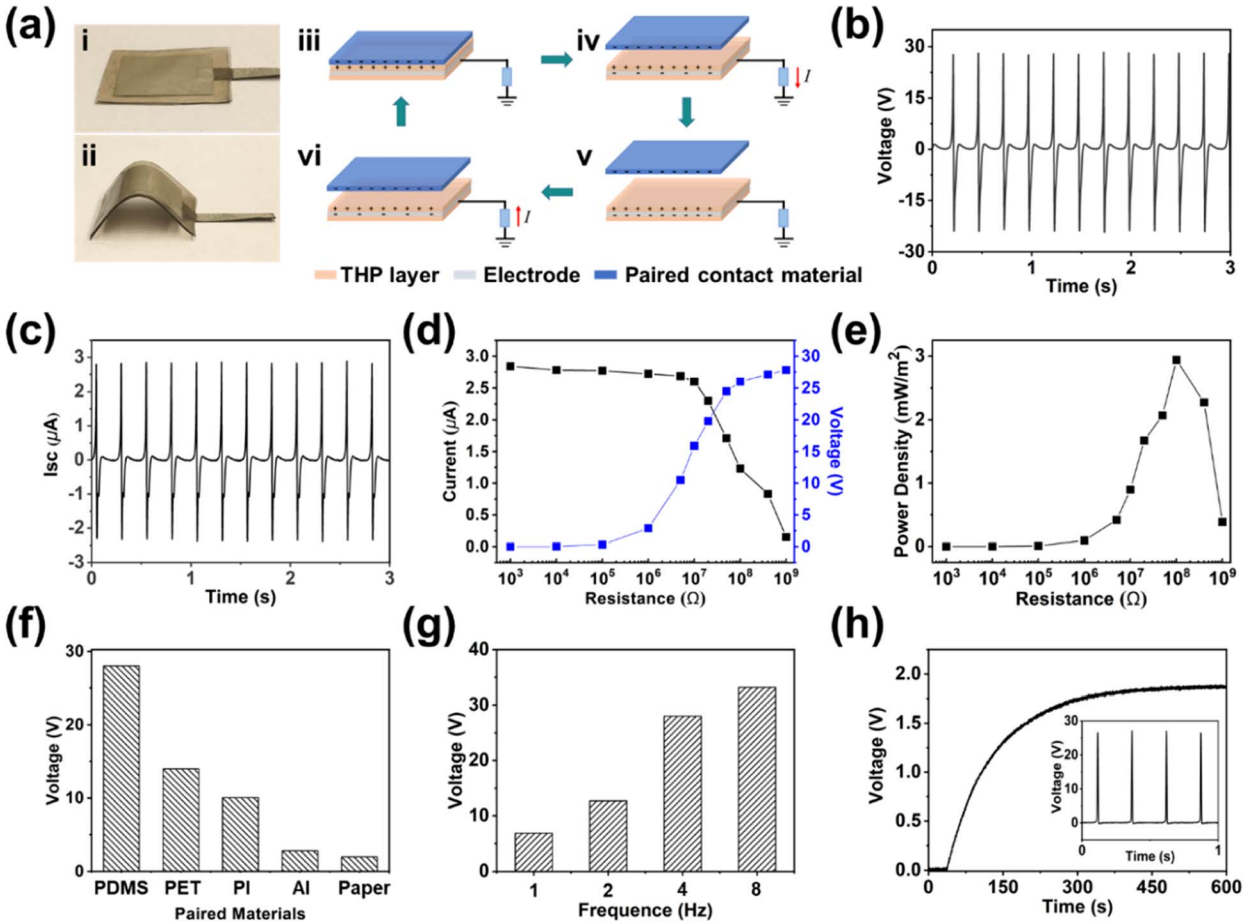

Fig. 3 The electrical performance of THP-TENG. (a) Photographs (i and ii) and the working principle (iii–vi) of THP-TENG. (b and c) Output voltage (b) and short-circuit current (c) of THP-TENG driven by a linear motor. (d) Output voltage and current with various resistances. (e) Output power density with various resistances. (f and g) Output voltage of the device under different paired contact materials (f) and contact frequencies (g). (h) The change in voltage of an 18 μF capacitor when charged by a THP-TENG, with the inset showing the DC electrical output of the THP-TENG combined with an inverter into the circuit.

previous reports.[15,50–53] Additionally, after introducing an inverter into the circuit, the AC signals can be converted to a DC electrical output for charging capacitors. The voltage of the capacitor (18 μF) reached about 1.87 V after being charged by a THP-TENG for 10 min as shown in Fig. 3h.

The prepared device can also serve as a self-powered sensor for detecting external stimuli. As shown in Fig. 4a(i), we firstly fabricated a THP-TENG with an array of square-shaped electrodes as a self-powered tactile sensor. It was found that when a finger slides on the surface of the device (*e.g.* the area in the red box in Fig. 4b), only those electrodes under the contact area (electrodes 1–3, Fig. 4b) generate significant electric signals as shown in Fig. 4c. This is because the triboelectric charges created from finger touching are localized and create maximum electrical output through the nearest electrode based on the electrostatic effect. Correspondingly, the THP-TENG can distinguish the location of external mechanical stimuli by measuring the electrical output signals of different electrodes. Furthermore, adding an additional pair of electrodes (Fig. 4a(ii)) at the bottom surface of the THP-TENG (Fig. 4a(i)) creates independent channels (Fig. 4a(iii)) for the measurement of electrophysiological signals.[54] To demonstrate the functionality, we attached these devices on the medial and lateral forearm of a health volunteer, respectively (Fig. S7†), for collecting electromyography (EMG) signals. When three distinct gestures are made, distinguishable filtered EMG patterns are obtained from the bottom skin-contact electrode pair of the devices (Fig. 4d). Correspondingly, the device shows the potential for gestures recognition. To our knowledge, few attempts have been made at constructing healable devices with both mentioned functions.

As shown in Fig. 5a, the healing of THP-TENG can be conducted by simply covering the wetted breakage area with a piece of scotch tape, which is very convenient and safe to operate in daily application. In addition to the healing of device structures, the recovery of conductivity of the electrode was observed. As shown in Fig. 5b(iii), the electrode was in a high-resistance state when it was cut apart, where the voltage across the electrode approximates to the open-circuit voltage (V_{oc}). Its conductivity recovered to the original value upon healing of the THP layer. Correspondingly, the extinct LED light (Fig. 5b(i)) connected in series with the electrode lit up again (Fig. 5b(ii)). Owing to the

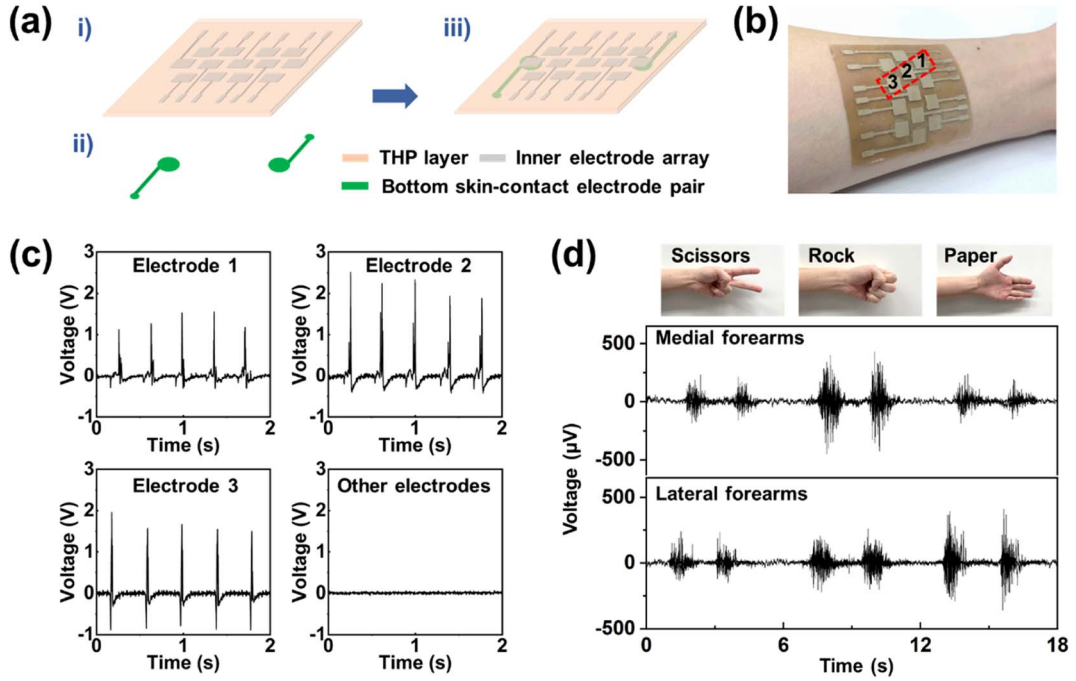

Fig. 4 The sensing performance of THP-TENG in detecting tactile and electrophysiological signals. (a) Schematics of a THP-TENG-based sensor including a square-shaped inner electrode array and circle-shaped skin-contact electrode pair on the bottom surface. (b) A photograph of the THP-TENG-based sensor attached on the medial forearm. (c) Electrical signals generated from different square-shaped inner electrodes when a finger slides on the surface of the device. (d) Three distinct gestures and the corresponding filtered EMG signals collected from the bottom skin-contact electrode pair of two devices attached on the medial and the lateral forearm, respectively.

recovery of the electrodes' conductivity, restoration of the electrical output of the THP-TENG is realized, and the output voltage of the healed device can recover to the initial value of about 28 V as shown in Fig. 5c. It was also found that the healed device can withstand long-term and continuous operation for 8000 cycles without performance degradation as shown in Fig. 5d. The THP-TENG also shows recyclability due to the solution processability of THP. As shown in Fig. 5e, the discarded device (i) can be broken into small pieces (ii) and then dissolved into DMF (iii). After centrifuging, the Ag electrode portion is separated from the system (iv), and the THP film can be formed again after evaporating the solvent of the

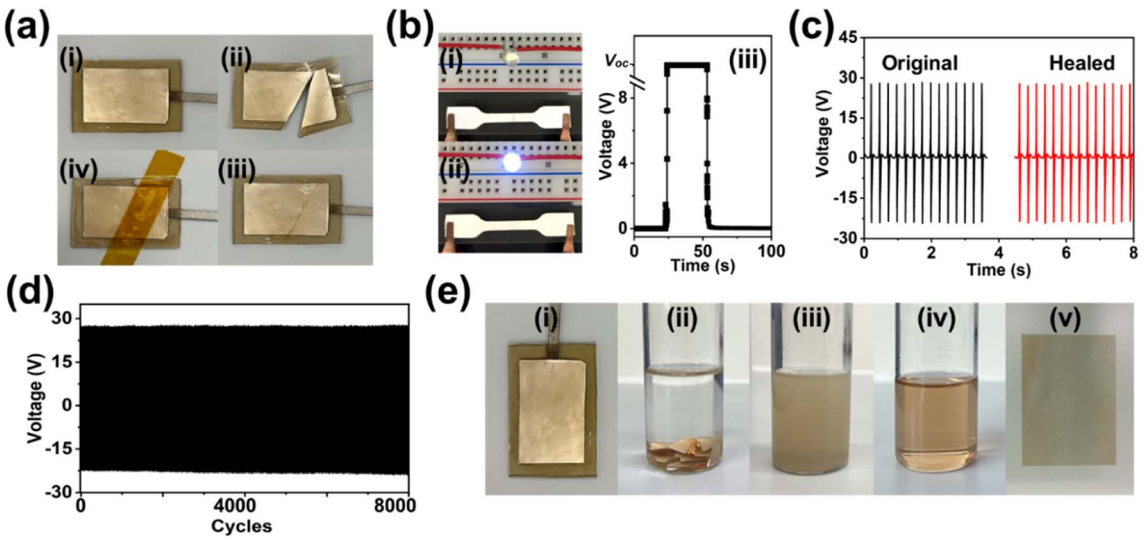

Fig. 5 The healing and recycling performance of the device. (a–c) Healing of the device integrity (a), electrical conductivity of the electrode (b), and the output voltage of the device (c). (d) The output voltage of the healed THP-TENG during continuous operation for 8000 cycles. (e) The recycling process of THP-TENG using DMF solvent.

supernatant (v). This recyclability makes the device economical and environmentally friendly for further application.

Conclusion

In this study, we developed a mechanically strong and mildly healable polymer, namely THP, as the base material of TENGs. The tensile strength of THP (C_{Ca} = 28%) can reach 11.6 MPa, and the healing of THP can be easily conducted with the assistance of water or alcoholic drinks with the healing ratio of about 90%. TENGs based on THP are mechanically robust and also healable under mild conditions which is convenient in daily operation. After healing the structural damage, the output voltage of the device can recover to the initial state, and the healed device shows stable performance during continuous operation for 8000 cycles without performance decay. In addition, the THP-TENG also possesses solution processability for device recycling. The prepared THP-TENG shows potential as the mechanical energy harvester and the self-powered sensor for detecting tactile and electromyographic signals, indicating possibilities for the development of advanced soft electronics with improved mechanical resilience.

Conflicts of interest

There are no conflicts to declare.

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