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Flexible piezoelectric nanogenerator in wearable self-powered active sensor for respiration and healthcare monitoring

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Abstract

A wearable self-powered active sensor for respiration and healthcare monitoring was fabricated based on a flexible piezoelectric nanogenerator. An electrospinning poly(vinylidene fluoride) thin film on silicone substrate was polarized to fabricate the flexible nanogenerator and its electrical property was measured. When periodically stretched by a linear motor, the flexible piezoelectric nanogenerator generated an output open-circuit voltage and short-circuit current of up to 1.5 V and 400 nA, respectively. Through integration with an elastic bandage, a wearable self-powered sensor was fabricated and used to monitor human respiration, subtle muscle movement, and voice recognition. As respiration proceeded, the electrical output signals of the sensor corresponded to the signals measured by a physiological signal recording system with good reliability and feasibility. This self-powered, wearable active sensor has significant potential for applications in pulmonary function evaluation, respiratory monitoring, and detection of gesture and vocal cord vibration for the personal healthcare monitoring of disabled or paralyzed patients.

Supplementary material for this article is available online

Keywords: piezoelectric nanogenerator, self-powered wearable active sensor, PVDF, respiratory monitoring, tachypnea

(Some figures may appear in colour only in the online journal)

Introduction

The human respiratory system is a series of organs responsible for taking in oxygen and expelling carbon dioxide. When a breath is taken, the diaphragm flattens out and pulls forward, making more space for the lungs, and during exhalation, the diaphragm expands and forces air out [1]. It is known that diaphragmatic breathing is the most natural way of breathing. In adult humans at rest, any rate between 12 and 20 breaths per minute is normal and tachypnea is indicated by a ventilatory rate greater than 20 breaths per minute [2]. Children have significantly higher resting ventilatory rates, which decline rapidly during the first three years of life and then more steadily until 18 years of age. Tachypnea can be an early medical sign of pneumonia in children [3]. Monitoring the diaphragmatic breathing can provide information about the movement of the diaphragm and is crucial for diagnoses

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of respiratory system disease, medical treatment observation, and postoperative care [4–7]. Most systems use strain gauges made from a mercury-filled silastic, a silicone elastomer that is extremely sensitive to movement, resulting in a lower signal-to-noise ratio. Non-invasive skin electrodes are placed on the thorax, and the variation of the electrical impedance can be detected during respiration cycles. However, these gauges are susceptible to humidity and electromagnetic environments. Other methods, such as nasal air flow sensors, are not reliable and therefore not commonly used for respiratory monitoring [8]. Pressure sensors based on a nanogenerator provide a new path for the monitoring of physiological signals [9].

In recent years, many different types of nanogenerators and sensors based on piezoelectric effects have been demonstrated, and many piezoelectric materials have been adopted. Ceramics such as lead zirconium titanate (PZT) [10, 11], barium titanate ($BaTiO_3$) [12, 13], and semiconductors such as zinc oxide (ZnO) [14–17] are the most widely used piezoelectric energy-harvesting materials. However, the low piezoelectric coefficient of ZnO impedes the development of high-output piezoelectric devices [18, 19], and although ceramics possess larger piezoelectric coefficients than polymers, they also possess higher elastic moduli and are hence stiffer than polymers, less sensitive to small deformations, and more prone to stress failure [20, 21]. Piezoelectric polymers with large piezoelectric coefficients such as poly(vinylidene fluoride) (PVDF) ($d_{31} \approx 20\,\mathrm{pC/N}$ and $d_{33} \approx -20$ to $-30\,\mathrm{pC/N}$) have attracted great attention [22]. Owing to its structural flexibility and processing simplicity, PVDF can be easily processed into light and flexible thin-film systems [18]. These are potentially superior materials as they are robust, lightweight, and easy and cheap to fabricate in addition to being lead free and biocompatible [23–25]

In this paper, a simple, durable, and cost-effective PVDF piezoelectric nanogenerator (NG) was fabricated. This flexible NG can be used as a wearable self-powered respiration sensor for real-time monitoring of human respiratory conditions and relaying of feedback. The cyclic expand–contract movement of the human body will lead to the stretching–releasing process of NG as respiration goes on. The relationship between the output electrical signals obtained and the respiration signals measured by a physiological signal recording system was thoroughly investigated. Subtle muscle movements in human gestures and vocal cord vibrations were also detected for personal healthcare monitoring.

Results and discussion

The detailed structure of an NG fabricated as a piezoelectric active sensor (PEAS) is shown in figure 1(a). It is primarily composed of three key components: the piezoelectric layer, electrodes, and elastic substrate. In this study, a PVDF film with an electrospinning PVDF structure was compressed and adopted as the piezoelectric layer because of its high piezoelectric voltage constant; its SEM image is shown in figure 1(c). Electrospinning has been demonstrated to be a unique method for the formation of submicron or nanofibers from polymer solutions, distinct from more traditional fabrication methods such as phase inversion. Electrospinning at lower chamber temperatures or fast evaporation of the solvent favor the formation of the β phase. In addition, the high voltage or high stretching ratio of the jets also benefits crystallization in the β phase.

The PVDF film is about 200 μm, on average, after compression. Gold film was deposited on both sides of the PVDF film as the top and bottom electrodes. The PVDF piezoelectric layer was initially polarized and all dipoles within are arranged in the direction of the external current electric fields at the original state (figure 2(a)). Silver paste was also applied to increase the adhesion between electrodes and lead wires. Silicone was employed as an elastic substrate to enhance the structural stability of the whole device. The PEAS showed great flexibility and deformation-recovery ability (figure 1(b)). The dimensions of the whole PEAS are 42 mm $\times$ 20 mm $\times$ 0.6 mm. When the PEAS was bent by a cyclic mechanical force (about 37 N, 1.4 Hz) from a linear motor, the generated open-circuit voltage and short-circuit current were around 1.5 V and 400 nA respectively, as illustrated in figures 1(d) and (e). The uniform output electrical signals show the good stability of the as-fabricated PEAS.

After connecting both sides of the PEAS with bandages, it can be dressed on the human body as a detector to monitor respiration signals from the belly or chest. According to the good flexibility of PVDF film and silicone substrate, the PEAS can fit well with the shape of different areas of human body. As the cycles of respiration proceed, the belly and the chest will expand and contract accordingly, thus leading to the stretching and releasing of PEAS.

The detailed working mechanism of the piezoelectric nanogenerator-based respiration sensor is illustrated in figures 2(b) and (c). When the chest and belly expand, the PVDF film is stretched and bent. The upper surface of the PVDF film endures a tensile stress, while the compressive stress appears on the lower surface of the PVDF film. The generated electrons will flow from the top electrode to the bottom electrode and generate an output electrical signal. When the chest and belly contract, the external force is withdrawn, and NG is released and will recover to the original state due to the flexibility of the PVDF film. The stress conditions on the upper and lower surface are fully reversed, and the electrons accumulated at the bottom electrode will move back to the top one and build up an opposite electric pulse. Electrons are driven back and forth through the external circuit with the force from human chest and belly to compress and release the PVDF film, thus contributing to cyclic alternating signals during human breathing.

It should be noted that only the PVDF in the β phase exhibits piezoelectric effects due to the all-trans configuration as illustrated in figure 2(d), and the β phase can be promoted by straining to many tens of percent, or via an electric-field-induced transformation under fields of order 1000 kV cm^{-1} at elevated temperatures [20]. The Fourier transform infrared (FTIR) spectrum of the PVDF exhibits strong vibration peaks

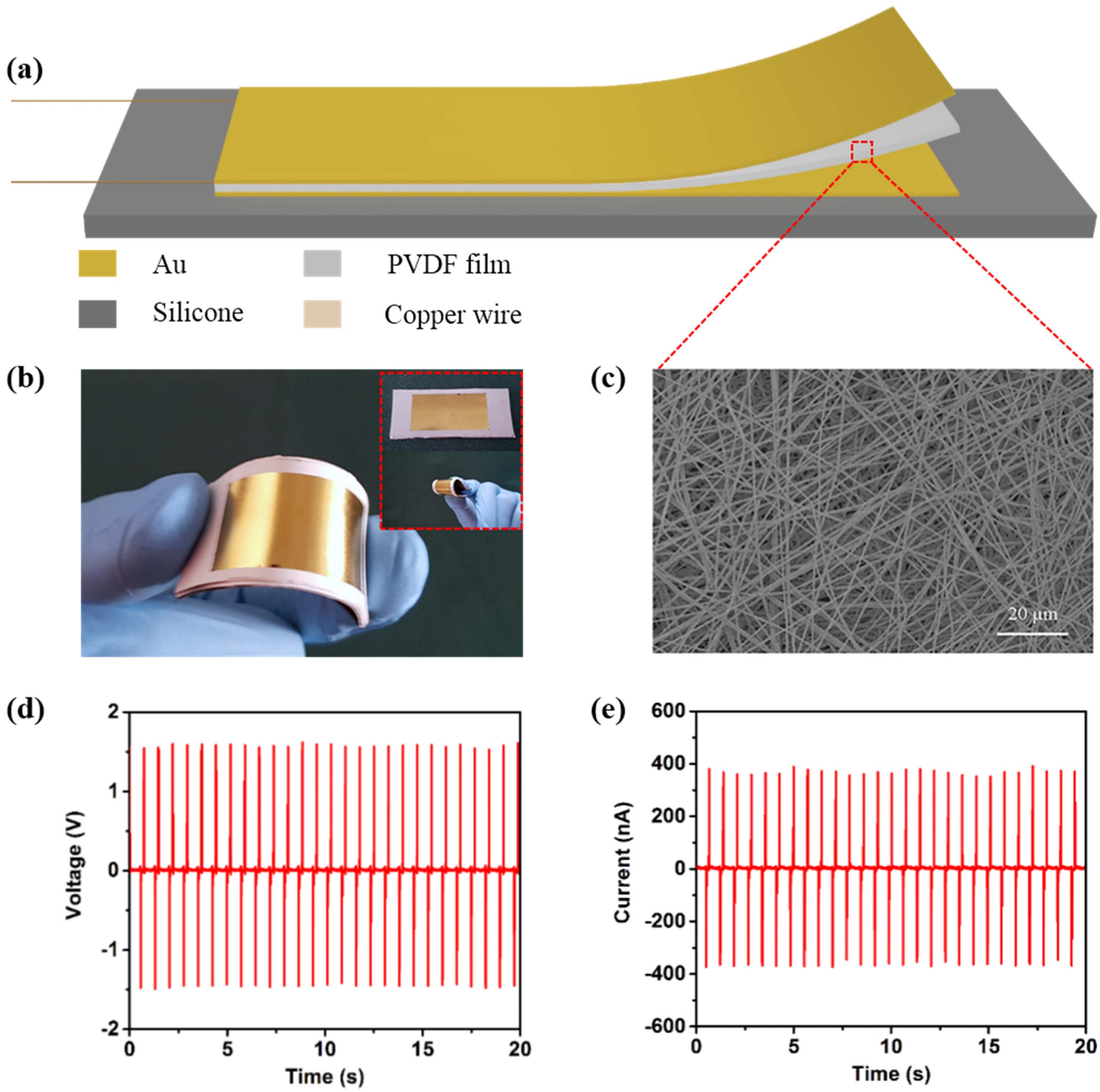

Figure 1. (a) 3D explosion view of the PEAS. (b) Photographs of PEAS based on PVDF NG. (c) SEM image of the PVDF electrospinning structure; the scale bar is 20 μm. The output electrical signals of the NG were characterized including the (d) open-circuit voltage and (e) short-circuit current.

at 510, 840, and 1280 cm^{-1} (figure 2(d)), which are typical for the b crystalline phase of PVDF.

The regular respiration frequency of a healthy adult is about 12–20 times in one minute, and if the number is beyond 20 bpm (beats per minute), it can be categorized as tachypnea. Tachypnea is a common clinical symptom of the respiratory system, and is often due to respiratory insufficiency caused by respiratory disease or respiratory organ or tissue lesions. If undetected, the disease will further aggravate the occurrence of respiratory distress, breathing difficulties, and even respiratory failure. Thus, it is of great importance to monitor the real-time human respiratory condition accurately if possible. Figure 3(a) is a schematic illustration of the working condition when the piezoelectric nanogenerator is applied to the human chest.

To demonstrate the efficiency and reliability of the device we proposed, we used the PVDF piezoelectric film-based PEAS and the physiological signal recording system Biopac MP150 to measure respiratory signals at different frequencies. The tested object was the respiration rate of a healthy male volunteer, who conducted his respiration rate at 18, 27, and 43 bpm respectively. The normal respiration rate of the volunteer was 18 bpm and, in order to imitate tachypnea, two faster frequencies (27 bpm and 43 bpm) of respiration were also adopted. To display the detailed signals more directly, we can compare the signals from both the PEAS and Biopac MP150. Respiration signals for about 20 s were shown in figures 3(b) and (c). The open-circuit voltage generated from PEAS was around 70 mV (figure 3(b)), and the respiration signals measured by the Biopac MP150 is shown in

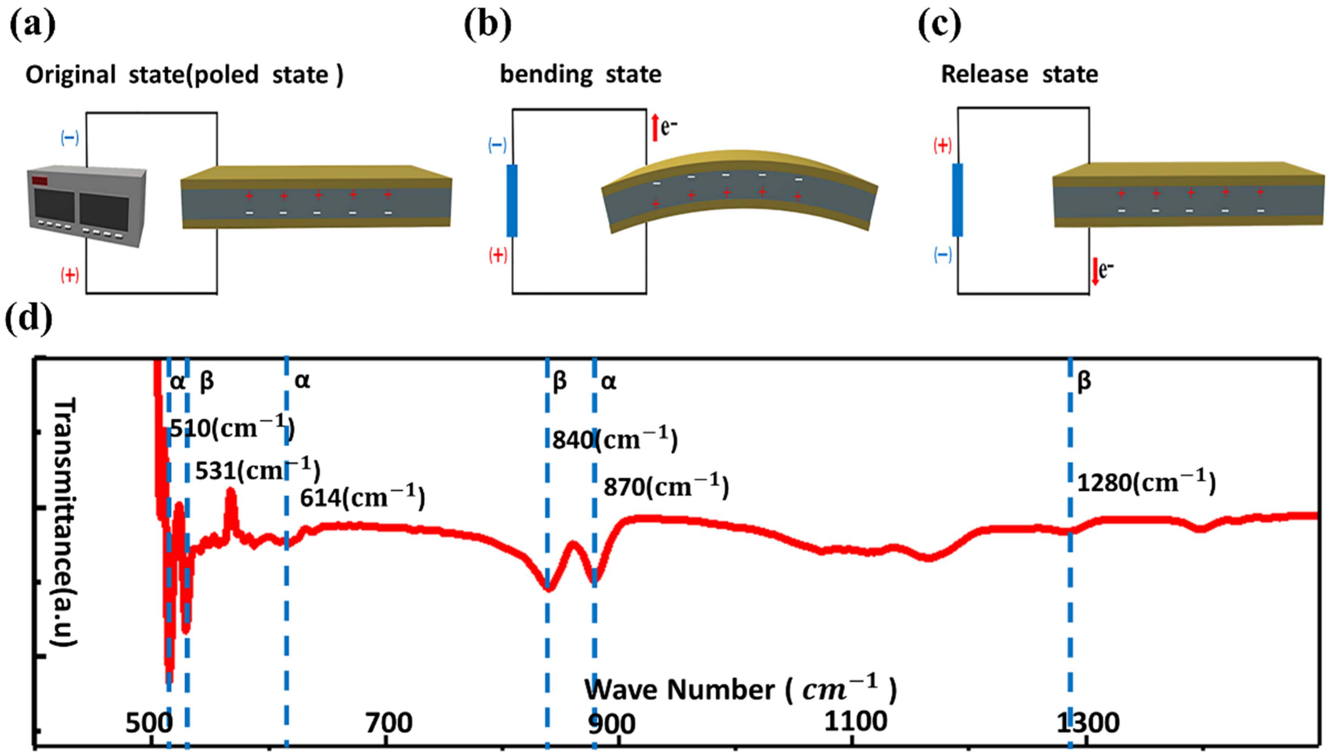

Figure 2. Schematics of the (a) polarization process, and (b), (c) working cycles of the piezoelectric nanogenerator-based respiration sensor. (d) FTIR spectra of the PVDF.

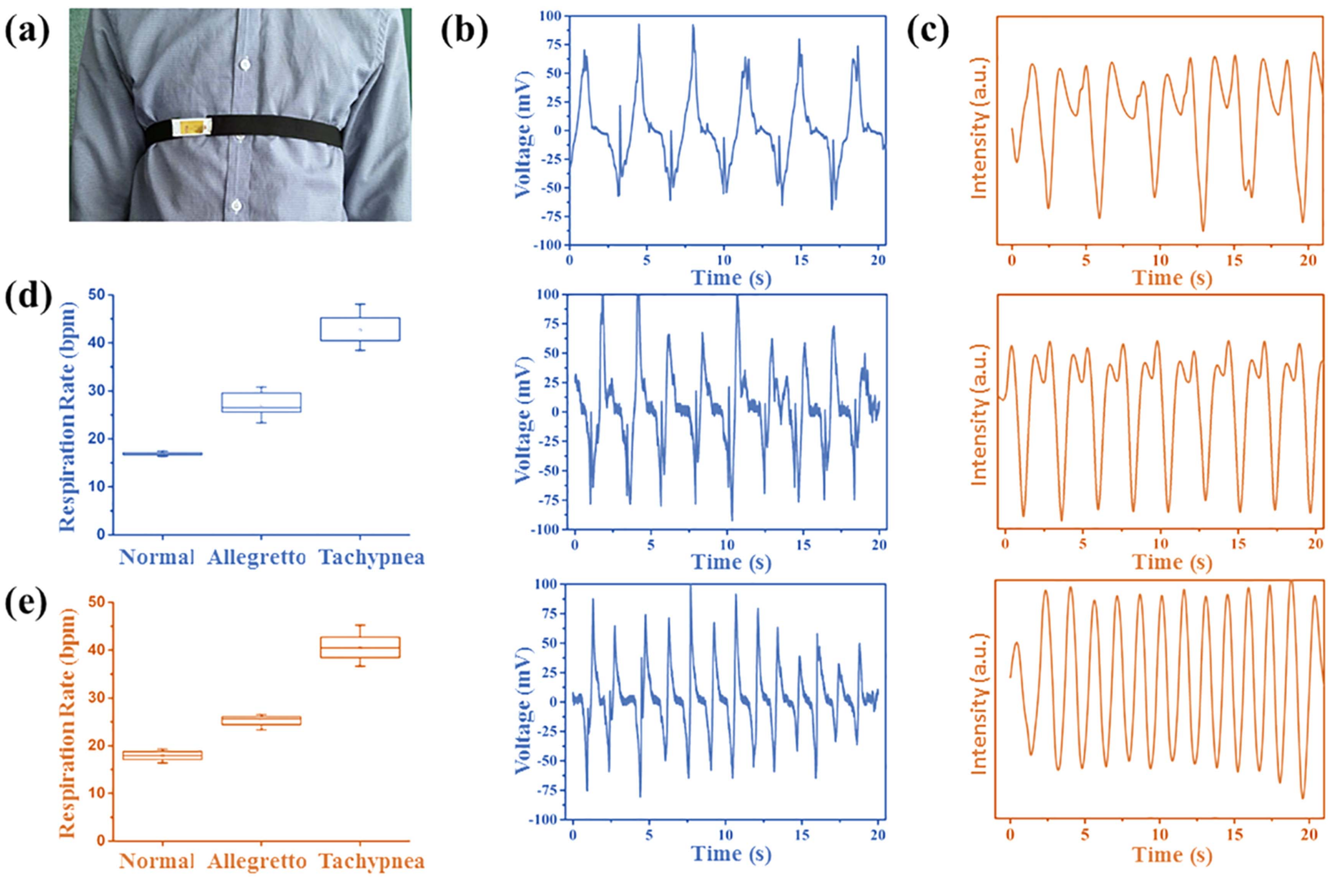

Figure 3. (a) Photograph demonstrating the PEAS as a wearable self-powered respiration sensor. (b) Open-circuit voltage of PEAS when breathing at different frequencies and (c) the corresponding respiration signals obtained from the physiological signal recording system (Biopac MP150). Statistical graph of (d) respiration rate of the piezoelectric nanogenerator and (e) Biopac MP150 results in different respiratory states.

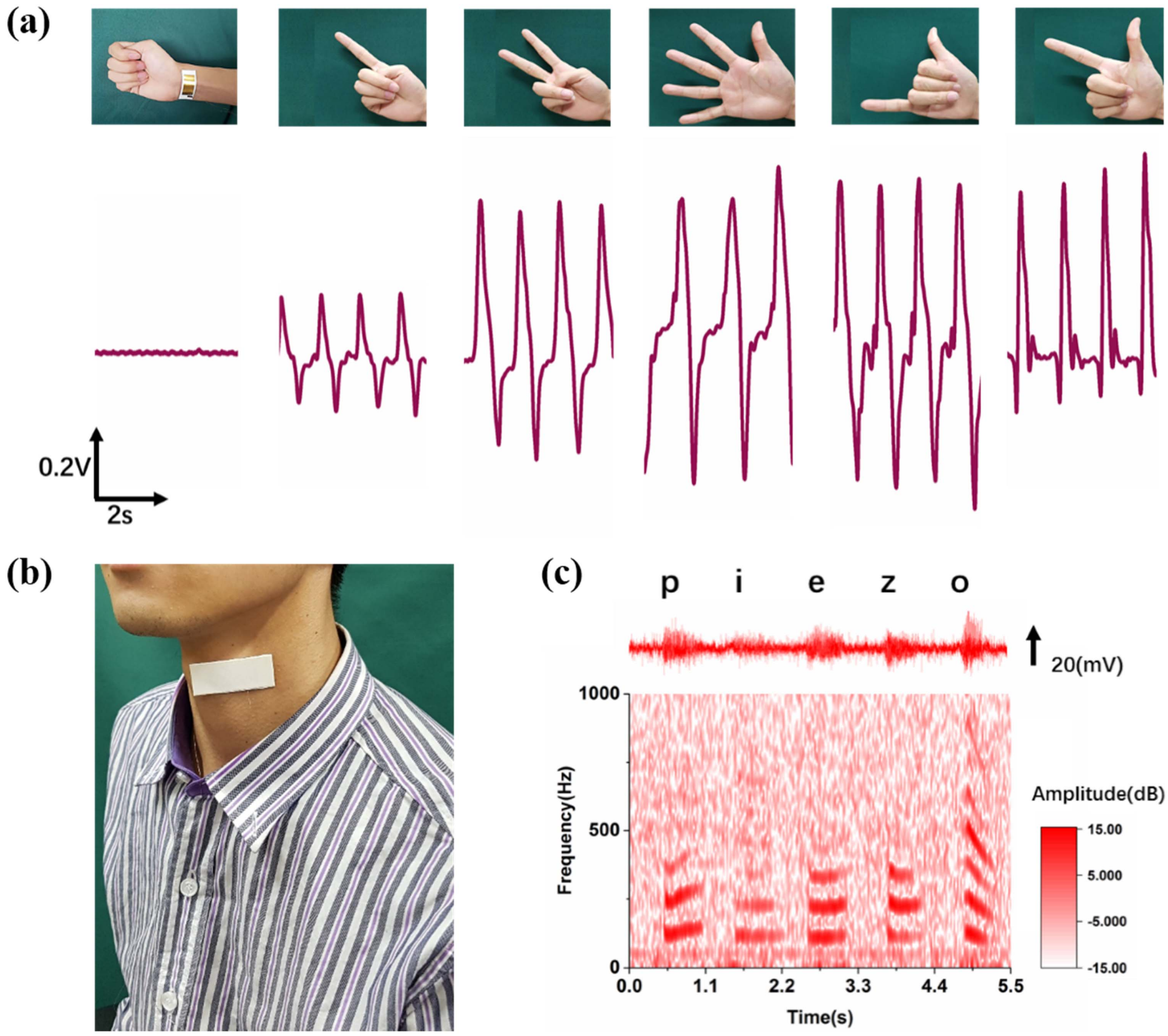

Figure 4. (a) Open-circuit voltage of the PEAS detecting subtle muscle movement based on different hand gestures. (b) Photograph showing the piezoelectric nanogenerator as an active sensor for human sound recording when attached to the skin of a volunteer's throat. (c) The open-circuit voltage signal when people spoke the letters 'P, I, E, Z, O' and the sonogram of the short-time Fourier spectrum of the signal.

figure 3(c). It can be seen clearly that the output electrical signals of the PEAS corresponds to the respiration signals measured by the Biopac MP150 (figures 3(b) and (c)). The respiration rate detected by the PEAS is the same as the values from the Biopac MP150 at three frequencies. Meanwhile, the curve of the PEAS is uncalculated and much more sensitive than the curves from the Biopac MP150 which have been calculated and modified in the physiological signal recording system. The sensitivity of PEAS is much more valuable for evaluating conditions such as interrupted respiration and respiratory distress syndrome.

The statistical graph of the respiration rate indicates that the PEAS (figure 3(d)) and Biopac MP150 (figure 3(e)) have high consistency at different respiratory rates. These results demonstrate the good sensitivity, reliability, and feasibility of the PEAS as a respiration sensor. These curves of respiration also contain detailed information about the dynamics of the respiration process. Quantitative diagnostic techniques may thus be developed in future on the strength of PEAS as a wearable and zero-power-consumption device.

To demonstrate the sensor's applicability to human healthcare monitoring, it was attached to a human wrist to detect movement of the underlying tendons and subtle muscle movement. The piezoelectric signal produced was measured based on different hand gestures (figure 4(a)). For the same gesture, the signals have good repeatability. Moreover, the signals have significant difference with different gestures, which shows a good resolution for tiny movements of the underlying tendons and arm muscle. This indicates a possible application of the PEAS in detecting subtle muscle movement in disabled and paralyzed patients for the control of healthcare equipment.

The PEAS demonstrated good ability to detect vibration of the vocal cords when it was attached to the skin of a volunteer's throat as an active sensor (figure 4(b)). No signal was observed when there was no speaking, corresponding to the silent state of vocal cords, while regular high-frequency signals emerged when the volunteer spoke the letters 'P, I, E, Z, O' separately. Figure 4(c) reflects the piezoelectric signals of the different letters. A sonogram was acquired by short-time Fourier spectrum using the signals generated from PEAS. The majority of the frequency components were distributed in the range of 50 to 1000 Hz when the volunteer spoke, as shown in the sonogram in figure 4(c). The frequency distribution demonstrates the sensor's good ability to detect vibration of vocal cords.

Conclusion

In summary, we have demonstrated a wearable, self-powered active sensor for respiratory sensing and healthcare monitoring that is based on a flexible piezoelectric PVDF nanogenerator. The open-circuit voltage and short-circuit current of the piezoelectric nanogenerator were characterized. The electrical signals of the sensor corresponded to the respiration signals measured by a physiological signal recording system. The stability and one-to-one relationship between the signals measured demonstrates that our fabricated wearable PEAS has the potential to sensitively monitor the human respiratory condition in real time. The PEAS can also detect human gestures and vocal cord vibrations. This suggests possible applications in the operation of healthcare equipment by detection of subtle muscle movements in disabled patients with exercise capacity loss or paralysis. As the PEAS is a zero-power-consumption device, we anticipate the technique demonstrated here can lead to the development of new quantitative diagnostic techniques for pneumology departments, and is of significant potential in developing healthcare sensors and devices.

Experimental and characterization section

Fabrication of PVDF film

The PVDF used was Foraflon® 4000HD. The solvents used were N,N-dimethylformamide and acetone. Solutions were prepared at concentrations of 15 wt% PVDF using as solvent pure DMF and a 3:1 v/v mixture of DMF with acetone. Solubilization was carried out at 70 °C under stirring for one hour.

A 20 ml glass syringe was used with a steel needle with internal diameter of 0.7 mm. The distance between the needle and collector was 3 cm, and the electric voltage applied was 10 kV. The collector used was an aluminum disk with diameter of 15 cm and width of 5 cm, at an angular velocity of 60 rpm.

Fabrication of flexible piezoelectric nanogenerator

The as-fabricated flexible piezoelectric nanogenerator is composed of the piezoelectric layer, electrodes, and elastic substrate. The 600 μm thick PVDF film was compressed to 200 μm on average, and adopted as the piezoelectric layer. 100 nm gold film was deposited on both sides of PVDF film by magnetron sputter as electrodes. The PVDF piezoelectric layer was polarized by external current electric fields of 5 KV. Silicone was employed as an elastic substrate to enhance the structural stability of the whole device. Two lead wires were connected to the gold electrodes by silver paste (figure 1(a)).

Fabrication of self-powered respiration sensor

The self-powered respiration sensor was fabricated by attaching a flexible piezoelectric nanogenerator to a stretchable bandage. This stretchable bandage can cincture the human chest (figure 3(a)) to detect respiration movement. The lead wires were protected at the edge of the bandage by Kapton tape without stretching.

Characterization methods

The microstructure of the PVDF was characterized by a scanning electron microscope (SU 8020). The open-circuit voltage of the piezoelectric generator pressed by linear motor was measured by Tektronix oscilloscope, and the open-circuit voltage generated by respiration movement was measured by Preamplifier SR570. The short-circuit current was measured using a Keithley 6514 system electrometer. Au film was deposited by magnetron sputter (Denton Discovery 635). The respiration signals were recorded by a physiological signal recording system (MP150, Biopac systems).

Human respiration monitoring

The self-powered respiration sensor was applied to the volunteer's chest. With the cyclic expand–contract movement of the human chest, the PEAS was stretched and released. The output electrical signals from the respiration sensor were obtained as respiration signals. The respiration signals of the volunteer were also detected for comparison and analysis by the physiological signal recording system Biopac MP150.

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