

A Breathable and Screen-Printed Pressure Sensor Based on Nanofiber Membranes for Electronic Skins

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In recent years, skin-like pressure sensors with high sensitivity and excellent flexibility are widely demonstrated for electronic skins. However, most of the reported skin-like pressure sensors are still based on airtight films, resulting in limited air permeability. Herein, cost-effective and capable processes of large-scale production are reported for lightweight and breathable pressure sensors based on nanofiber membranes (NM). The pressure sensor is composed of a layer-by-layer structure of poly(vinylidene fluoride) NM for substrates, silver nanowires for electrodes, and thermoplastic polyurethane NM for the dielectric layer through screen printing and ultrasonic bonding techniques. Benefiting from the high porosity of NM, the capacitive pressure sensor possesses unique performance, including a superior sensitivity of 4.2 kPa^{-1} , a fast response time ($<26 \text{ ms}$), an ultralow detection limit (1.6 Pa), and excellent breathability (Gurley value = $17.3 \text{ s}/100 \text{ mL}$). Furthermore, the pressure sensor is not only applicable to monitor human physiological signals, but also to detect spatial pressure distribution. These results indicate that the breathable and screen-print pressure sensor is promising for electronic skins with air permeability.

Promoted by the urgent demand for electronic skins (e-skins) which have great potential in artificial intelligence, wearable devices, and smart robots, the development of skin-like pressure sensors has attracted unprecedented attentions.^[1–10] Although skin-like pressure sensors with high sensitivity and good flexibility have been investigated, lightweight, breathable, and large-area pressure sensors, which could be applied in continuous physiological detection over a long period of time, are still highly desired.^[11,12] In order to fully mimic unique properties of human skin, the skin-like pressure sensors with transparency,^[3,13]

stretching,^[14,15] and self-healing^[16,17] have been widely demonstrated. Due to the airtight films, most of reported skin-like pressure sensors are not permeable for air, thus skin cannot breathe when attached to human body, which can result in inflammation after wearing for a long time.^[18] Therefore, pressure sensors with air permeability are crucial to skin-like electronics. Paper, which is made up of randomly interconnected micrometer cellulose fibers, offers a naturally porous structure.^[19] It could be widely used for the breathable substrates.^[20] However, the rough surface of the paper usually needs mineral fillers (e.g., calcium carbonate, chalk and clay) to increase smoothness for printed electronics, which weaken its flexibility and increase its density.^[21] Nanofiber membranes (NM) by electrospinning with inherently high porosity, excellent flexibility, and smoothness could be more potential in lightweight, breathable, and printable electronic.^[22]

Herein, we report a breathable and screen-print capacitive pressure sensor for electronic skins, which is composed of two poly(vinylidene fluoride) NM/silver nanowires (PVDFNM/AgNWs) electrodes and a thermoplastic polyurethane NM (TPUNM) dielectric layer. The skin-like pressure sensor with high sensitivity, good flexibility, air permeability, and light weight could be fabricated through low-cost and large-scale processes. PVDFNM with hydrophobicity and breathability is prepared for substrates through electrospinning. The PVDFNM/AgNWs electrodes are fabricated through screen printing. Importantly, in comparison with the traditional microstructured polydimethylsiloxane (PDMS) dielectric layer, the electrospinning process of TPUNM is cost-effective and capable processes of large-scale production. The pressure sensors possess unique performance, including superior sensitivity of 4.2 kPa^{-1} , fast response time ($<26 \text{ ms}$), an ultralow detection limit (1.6 Pa), and excellent breathability (Gurley value = $17.3 \text{ s}/100 \text{ mL}$). Based on its unique performance, the flexible capacitive sensor is not only applicable to monitor human physiological signals, but also to realize a spatially resolved sensing by print-integrated sensor arrays.

The design of breathable pressure sensor is based on the structure of the parallel plate capacitor with two PVDFNM/AgNWs electrodes and a TPUNM dielectric layer, as shown in Figure 1a. Figure 1b–d shows the scanning electron microscope (SEM) images of the surface of the porous PVDFNM for

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DOI: 10.1002/admt.201700241

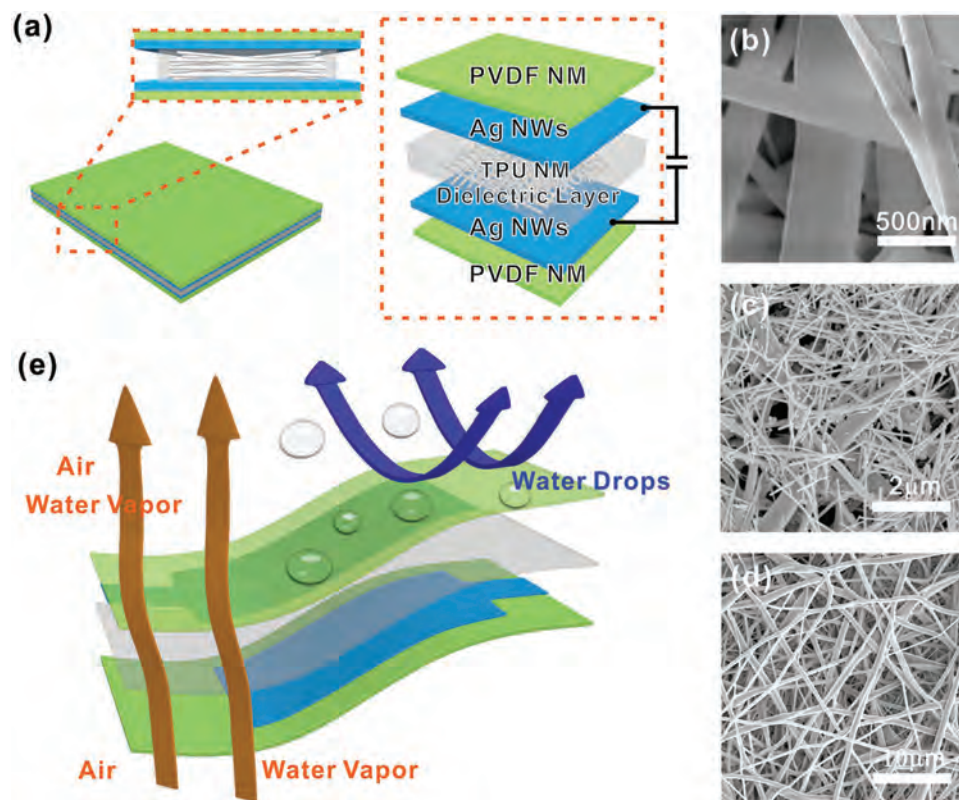


Figure 1. Structure and function of the breathable skin-like capacitive pressure sensor. a) Structure of the skin-like pressure sensor based on NM. b) Porous surface of the PVDFNM in SEM. c) Interlaced AgNWs on the surface of the PVDFNM in SEM. d) Porous surface of the TPUNM. e) The illustration of waterproof and breathable property of the sensor.

substrates, interlaced AgNWs adhered to the surface of the PVDFNM for electrodes, and porous TPUNM for dielectric layer, with many gaps ranging from tens of nanometers to several micrometers. These gaps are larger than the size of air molecules.^[23] Besides, PVDF is a kind of polymer with inherently good hydrophobicity and excellent biocompatibility.^[24,25] The surface contact angle of PVDFNM is 110.25° (Figure S1, Supporting Information), ensuring the good hydrophobicity of breathable pressure sensor. As shown in Figure 1e, the pressure sensor is impermeable to water and permeable to air. These characters of breathable pressure sensor are similar to human skin. The PVDFNM substrates can endow the pressure sensor with good biocompatibility. To evaluate its air permeability and physically biocompatibility, the breathable pressure sensor and nonbreathable PDMS film were attached to the forearm for 24 h (Figure S2a, Supporting Information). There are allergic reactions in the area of PDMS film. However, no allergic reactions have been founded in the area of breathable pressure sensor (Figure S2b, Supporting Information).

The fabrication processes of the pressure sensor are shown in Figure 2. The preparation of PVDFNM/AgNWs electrodes is accomplished through electrospinning and screen printing (Figure 2a–c). Compared with traditional fabrication approaches such as multistaged photolithography and vacuum deposition, these fabrication processes with high throughput and low-temperature processing have various advantages including high efficiency, simple operation, little waste, low-

cost, and large-scale production.^[9,26] Seen from the SEM image (Figure 1b), the surface of PVDFNM composed of nanofibers is smoother than the paper composed of microfibers, which is suitable for printing. The AgNWs were synthesized by polyol reduced processes.^[27] The diameter of the AgNWs is about 50–200 nm and the lengths can reach to tens of micrometers (Figure S3a, Supporting Information). The AgNWs screen-print inks are obtained by dissolved the AgNWs in ethylene glycol, ethylene glycol butyl ether, and hydroxypropyl cellulose mixture solution. The contact angle of as-prepared AgNWs inks on the surface of PVDFNM is 26.6° (Figure S3b, Supporting Information), ensuring good adhesion in printing. As for dielectric layer, the porous TPUNM instead of the microstructured PDMS is obtained by electrospinning technology (Figure 2a,b,d). The micro-electromechanical system fabrication process of microstructured mold of PDMS is complicated and expensive. In comparison, the fabrication process of TPUNM is cost-effective and capable processes of large-scale production. The pressure sensor is face-to-face assembled by bottom and top PVDFNM/AgNWs electrodes and middle TPUNM dielectric layer through ultrasonic bonding (Figure 2e,f).

The sensing performance of capacitive pressure sensors has been analyzed and the corresponding results are shown in Figure 3. The performance of pressure sensors based on TPUNM with various electrospinning times were investigated (Figure 3a). These sensors were denoted as TPUNM1, TPUNM2, and TPUNM3, corresponding to electrospinning

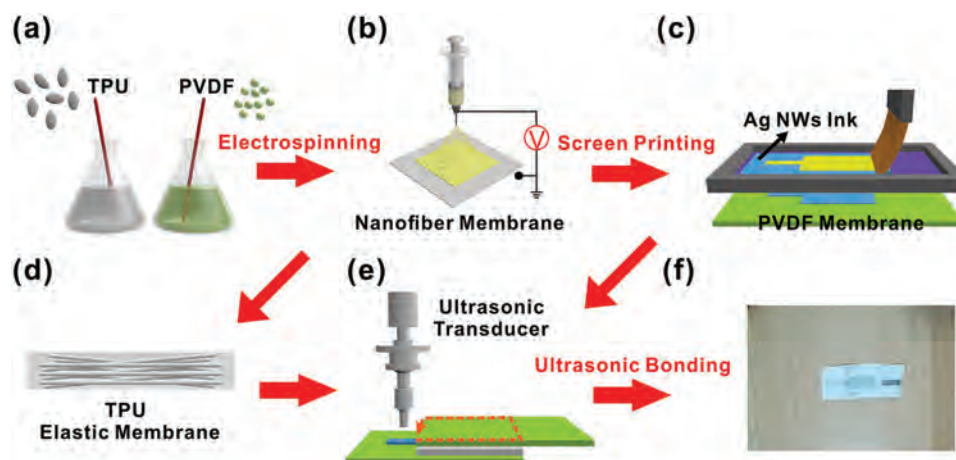


Figure 2. Fabrication process of the breathable skin-like capacitive pressure sensor. a) The precursor solution TPU or PVDF. b) The process of electrospinning of TPUNM or PVDFNM. c) Fabrication of breathable AgNWs/PVDFNM through screen printing. d) Schematic illustration showing elastic TPUNM. e) Fabrication of the breathable sensor through ultrasonic welding. f) Schematic illustration showing the skin-like pressure sensor.

time of 6, 4.5, and 3 h, respectively. The average densities of the TPUNM1, TPUNM2, and TPUNM3 are 0.379, 0.306, and 0.242 g cm^{-3} , respectively. Figure 3a shows the relative capacitive change of the TPUNM pressure sensors when external pressure is applied. The sensitivity is defined as $S = (\Delta C/C_0)/P$, where ΔC and C_0 present the variation of capacitance and the initial capacitance, respectively, and P presents the applied pressure (F/A , where F is the force applied on the pressure sensor and A is the stressed area of the pressure sensor). Pressure sensors with thinner TPUNM exhibit higher sensitivity. For example, the sensitivities of TPUNM3 are 4.2 kPa^{-1} for the low-pressure range of 0–400 Pa, and 0.071 kPa^{-1} for the high-pressure range of 4–30 kPa. In comparison, the sensitivities of TPUNM1 are 3.3 and 0.059 kPa^{-1} . With the decreasing of electrospinning time, the density of TPUNM is lower. The lower density of TPUNM3 results in the higher relative distance change during the pressure applying, thereby improving the sensitivity of pressure sensor. Remarkably, the pressure sensor can achieve higher sensitivity than one based on microstructured PDMS dielectric layer.^[28] Many air gaps exist in the porous structure of TPUNM, providing a larger amount of deformation space for the dielectric layer, which is critical for sensitivity enhancement. To investigate the response time of the breathable pressure sensor, dynamic pressure has been applied on the sensor. The response time of breathable pressure sensors based on NM is less than 26 ms (Figure 3b). PVDFNM substrate with high breathability can improve the air flow velocity in the dielectric layer, thereby accelerating the response speed of the pressure sensor. Besides, the capacitive change of pressure sensor is less than 0.01 pF without pressure in normal environment so that it can detect a pressure of 1.6 Pa with obvious change value of larger than 0.04 pF (Figure S4, Supporting Information). In comparison with the previous reports,^[29–34] the pressure sensor has higher sensitivity and lower detection limit (Figure 3c and Table S1, Supporting Information). In order to evaluate its breathability, the Gurley values of PVDFNM, PVDFNM/AgNWs electrode, TPUNM dielectric layer, and whole pressure sensor are measured, and the Gurley values (Gurley value presents the time of 100 mL air to pass through the sample, and it reflects the

difficulty of air to pass through the membrane, which is widely used in measuring the breathability of thin films) of these samples are 4.9, 8.1, 1.3, and 17.3 s, respectively (Figure 3d). The air permeability of pressure sensor is attributed to the porous structure of NM. Furthermore, the breathable pressure sensor exhibits excellent reproducibility and durability without obviously change in 10 000 cycles at a frequency of 1.43 Hz (0.7 s for one cycle) under pressure of 15 kPa (Figure 3e). These results suggest the skin-like pressure sensors could be applied in the wearable and breathable health-monitoring devices.

Although flexible pressure sensors have been widely used in wearable health-monitoring devices,^[35] their air impermeability has restricted the further practical application in continuous physiological detection over a long period of time due to inflammation of skin.^[18] Our pressure sensor with air permeability could be more potential in monitoring human physiological signals, such as respiration and heart rate (HR). A smart mask with breathable sensing equipment to detect breathing conditions is particularly important. Namely, our pressure sensors with air permeability are suitable for the mask. The physical force of a breath can be monitored in real time after attaching the sensor directly on the mask (Figure 4a and Movie S1, Supporting Information). According to the obtained results, the respiratory rates under normal condition and after exercise are 17 and 46 times per minute, respectively (Figure 4b). Besides, the influence of humidity ranging from 20% to 80% on the performance of sensor has been investigated (Figure S5 and Table S2, Supporting Information). And we also have measured the influence of low temperature ranging from 0 to 20°C on the performance of sensor (Figure S6 and Table S3, Supporting Information). Although the dependence of output capacitive on humidity and temperature can degrade the absolute performance pressure sensor, the capacitance or relative dielectric constant does not fluctuate clearly with changes in humidity or temperature. The TPUNM dielectric layer is hydrophobic so that this moisture cannot accumulate substantially on this membrane to form a water layer. Thus, when the man breathing is constantly monitored for a while, the capacitance and relative dielectric constant demonstrates tiny changes (Figure S7 and

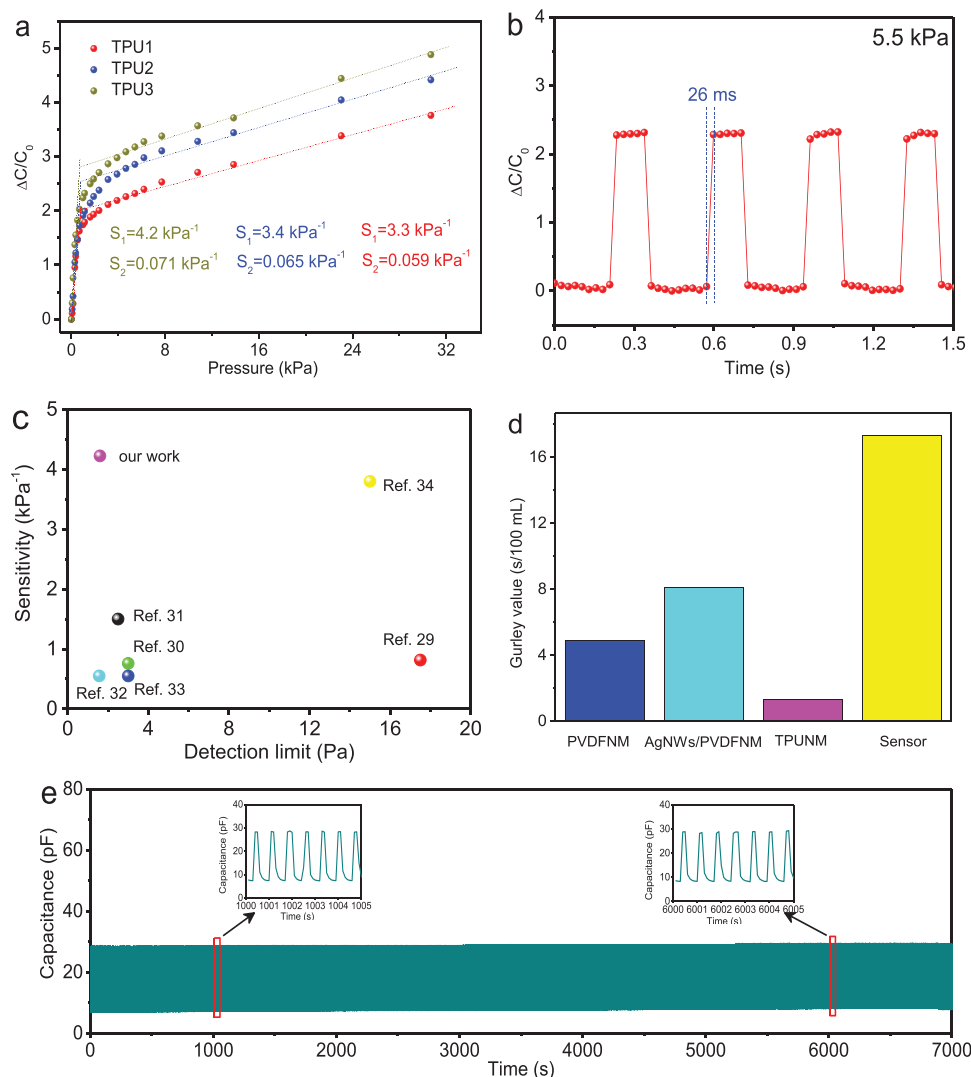


Figure 3. Performance of flexible pressure sensors. a) The performance comparison of TPUNM dielectric layers with different electrospinning time. b) The response time of breathable pressure sensor under an applied pressure of 5.5 kPa. c) Sensing performance of pressure sensors in previous reports. d) The Gurley value of PVDFNM, AgNWs/PVDFNM, TPUNM, and sensor. e) The cycling test of the breathable pressure sensor over 10 000 loading–unloading cycles at a frequency of 1.43 Hz under pressure of 15 kPa.

Table S4, Supporting Information). In addition, monitoring HR signs for long periods of time is important for assessing the physical and mental state of the human body. The air permeability of skin-like pressure sensors makes them suitable for HR monitoring. Our pressure sensors could be attached onto the chest to monitor HR (Figure 4c). The HR could be read out accurately under both normal condition (60 beats per minute) and after physical exercise (97 beats per minute), as shown in Figure 4d. These results demonstrate that the breathable skin-like pressure sensors are promising for subtle force, indicating its potential to serve as a wearable health-monitoring device.

In order to demonstrate the large-area pressure sensors for electronic skins, we have fabricated a 100 pixels pressure sensor matrix array to measure the pressure distribution through low-cost and large-area processes. The sensor matrix is comprised of bottom and top patterned PVDFNM/AgNWs electrodes through screen printing and middle TPUNM dielectric layer through

electrospinning. The breathable sensor with 10×10 pixel array and corresponding electronic device is shown in Figure 5a. When a heart-shaped metal object was positioned over the top of our sensor array (Figure 5b), the color contrast mapped local pressure distribution in consistency with the shape of the heart (Figure 5c). The metal letters “C,” “A,” and “S,” which are the logogram of the “Chinese Academy of Sciences,” were positioned on the top of sensor array (Figure 5d–f), and the pressure distributions were disclosed through current mapping of the sensor arrays. The local pressure distribution can be indicated by the color contrast, which is consistent with the shape of the three letters (Figure 5g–i).

In summary, we have demonstrated a skin-like pressure sensor with high sensitivity, good flexibility, fast response speed, outstanding air permeability, and light weight through electrospinning and screen printing, which are suitable for low-cost and large-area production. Due to many air

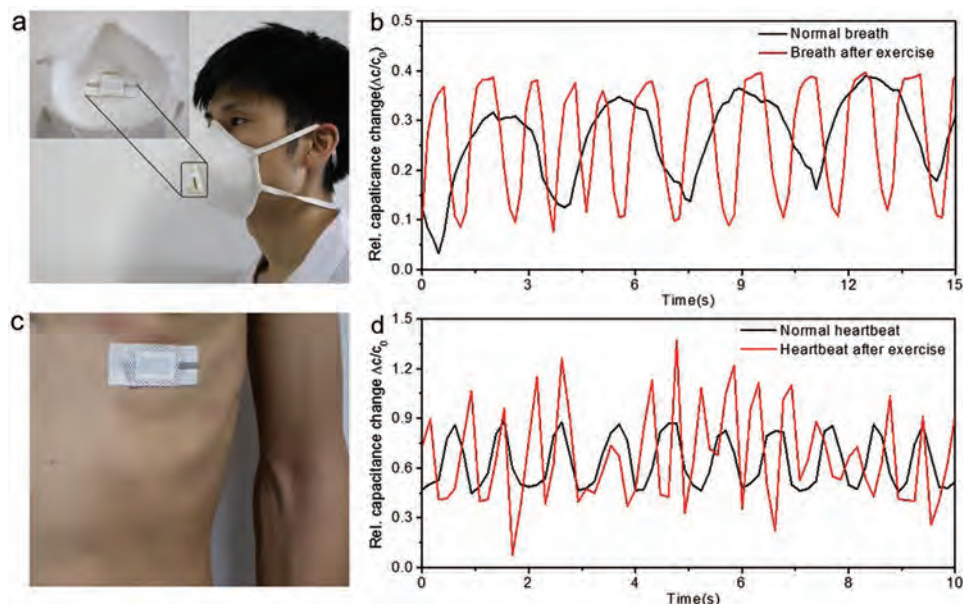


Figure 4. Breathable pressure sensor for human physiology monitoring. a) The sensor attached to the mask for respiration monitoring. b) The result of respiration under normal and exercise conditions. c) The sensor attached to the chest for heart rate. d) Signal of the physical force of heart rate under normal and exercise conditions.

gaps of TPUNM, the skin-like pressure sensor exhibited high sensitivity of 4.2 kPa^{-1} . Based on good air circulation in the dielectric layer, the pressure sensor has shown fast response

time of less than 26 ms. Besides, the skin-like PVDFNM substrate has endowed the pressure sensor with air permeability and hydrophobicity. The skin-like sensor was applied in

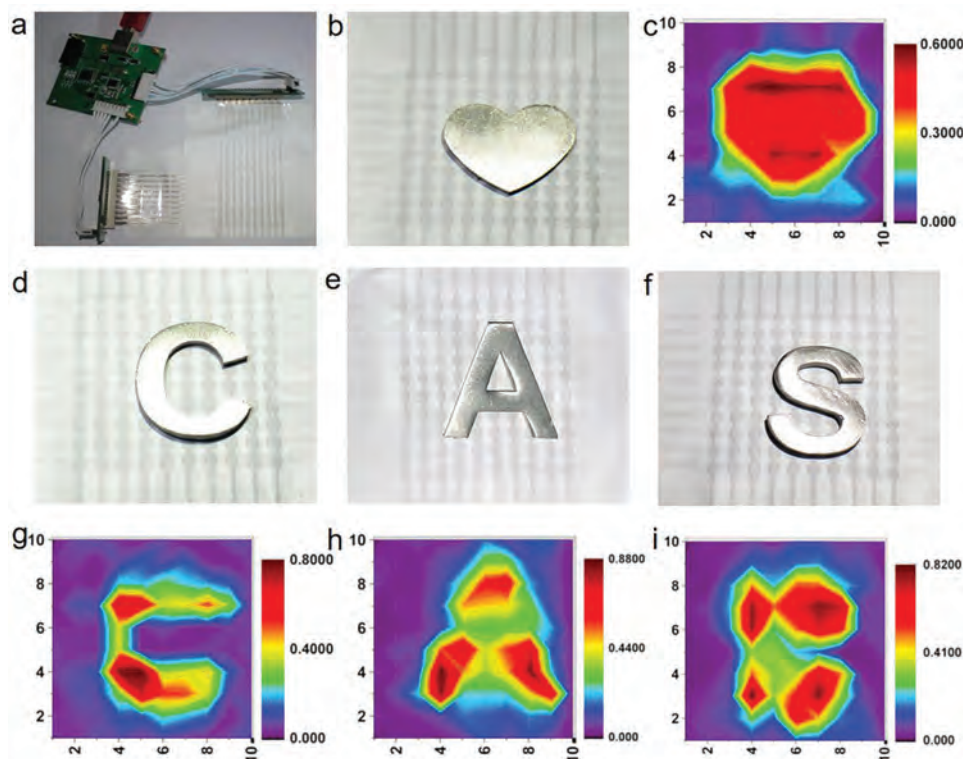


Figure 5. Photograph and performance of breathable pressure sensor array. a) Photograph of a flexible 10×10 pressure sensor array. b) Top view of the "heart" and c) the corresponding pressure signal distribution. d–f) Top view of the "C," "A," and "S" letters positioned over the pressure sensor array and g–i) the corresponding pressure signal distribution.

monitoring human physiological signals such as respiration and HR. In addition, a large-area pressure sensor array has been fabricated by screen printing and realized the detection of the external pressure distribution. As a result, the breathable and screen-print capacitive pressure sensor could provide a promising strategy for the design of e-skins with air permeability.

Experimental Section

Preparation of AgNWs and AgNWs Screen-Print Inks: The AgNWs were synthesized by polyol process. First, 3.03 g polyvinyl pyrrolidone (PVP) was dissolved in 253.8 g ethylene glycol (EG) by stirring at 160 °C for 2 h in a flask. Then 480 mL of 0.2 M NaCl in EG solution was added into the flask. After 2 min, 2.98 g EG solution containing 3.01 g AgNO₃ was added to the flask at the rate of 100 μ L/10 s. When the reaction solution became glistening, the reaction was stopped immediately. The obtained AgNWs were washed by acetone and deionized water to remove the residual PVP. AgNWs were collected. Ethylene glycol, ethylene glycol butyl ether, and hydroxypropyl cellulose were mixed at the weight ratio of 2:1:0.1 with 600 rpm magnetic stirring for 2 h to make additive solution. Finally, the additive solution was added to the AgNWs to obtain AgNWs screen-print inks.

Fabrication of PVDFNM and TPUNM: First, PVDF was dissolved in dimethylformamide (DMF) and magnetic stirred for 3 h until a homogenous to obtain 10 wt% PVDF precursor solution. Then, a high electric potential of 20 kV was applied at the tip of the syringe needle and the feed rate of PVDF solution was fixed at 0.6 mL h⁻¹ by using a flow-metering pump. The collecting distance between needle tip of syringe and the collector was 15 cm. A stainless steel drum substrate was used as a collector. After electrospinning for 6 h and dried at 70 °C for 30 min, the PVDFNM was obtained. For the TPUNM, the precursor solution was 20 wt% TPU in DMF and dichloromethane methylene chloride mixture solution. The other spinning conditions were the same as that of PVDFNM.

Design and Fabrication of the Skin-Like Pressure Sensor: The AgNWs ink was printed on the PVDFNM through screen printing. After drying at 60 °C for 3 h, the patterned PVDFNM/AgNWs electrode was obtained. The middle TPUNM dielectric layer and bottom and top patterned PVDFNM/AgNWs electrodes were face-to-face assembled into a capacitive pressure sensor through ultrasonic bonding. For flexible pressure sensor matrix fabrication, AgNWs pixels with a size of 2 \times 2 mm² were printed on the PVDFNM substrate for a 10 \times 10 array with a total area of 4 \times 4 cm². Two PVDFNM with patterned AgNWs matrix and a TPUNM dielectric layer were assembled into a skin-like capacitive pressure sensor matrix.

Characterization of the Skin-Like Pressure Sensor: SEM images were carried out using a Nova NanoSEM 450 at a 5 kV beam voltage. The contact angle measurement was measured using an XG-CAM Contact Angle Meter. The air permeability was performed using a Gurley-type densometer (4110N, Gurley) by measuring the time of 100 mL air to pass through the sample. Capacitance was characterized using a Keysight E4980AL Precision LCR meter at 1 kHz frequency and 1 V ac signal. Pressure was applied using a mechanical performance testing system (Micro Tester, 5848, Instron). For cyclic loading–unloading of pressure, a preselected constant pressure was repeatedly applied and released on the pressure sensor using a linear motor system.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

Acknowledgements

W.Y., N.-W.L., and S.Z. contributed equally to this work. The authors wish to thank Linlin Li, Shu Wang, and Chaoying Zhang for assistance on characterization measurements. This work was supported by the National Key R&D Project from the Minister of Science and Technology, China (2016YFA0202702, 2016YFA0202703), the Natural Science Foundation of China (NSFC Nos. 21274006, 21703010, and 51503005), and the Programs for Beijing Science and Technology Leading Talent (Grant No. Z16111000490000).

Conflict of Interest

The authors declare no conflict of interest.

Keywords

air permeability, electronic skins, nanofiber membranes, screen printing, skin-like pressure sensors

Received: September 5, 2017

Revised: October 11, 2017

Published online:

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