



Research progress of flexible wearable pressure sensors

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ARTICLE INFO

Article history:

Received 6 February 2021

Received in revised form 30 April 2021

Accepted 13 May 2021

Available online 18 May 2021

Keywords:

Piezoresistive pressure sensors

Capacitive pressure sensors

Piezoelectric pressure sensors

Self-powered

Multifunctional

Self-healing

ABSTRACT

Flexible pressure sensors with high sensitivity, high flexibility, lightness and easy integration have been extensively researched in the fields of electronic skin, wearable devices, medical diagnosis, physical health detection and artificial intelligence. **This review summarizes the latest research progress of piezoresistive pressure sensors, capacitive pressure sensors, and piezoelectric pressure sensors.** In addition, high-performance flexible pressure sensors designed for different application requirements such as self-powered pressure sensors, multifunctional pressure sensors, and self-healing pressure sensors are also discussed. After a comprehensive description of the latest flexible pressure sensors, we discussed the current challenges and potential prospects of flexible pressure sensors. Exploring new sensing mechanisms, seeking new functional materials, and developing novel integrated technologies for flexible devices will be the key direction in the sensor field in the future.

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1. Introduction

Flexible pressure sensors can bring important information about specific needs within the human body and during contact between the human body and the external environment [1]. Traditional semiconductor and metal-based pressure sensors are limited by their rigidity, fragility, low sensitivity, narrow sensing range, limited tensile capability and low resolution, making it difficult to apply them in applications requiring flexible contact or wearable devices. Therefore, people have explored flexible pressure sensors suitable for arbitrary curved surfaces, and have important significance in the fields of human-machine interface [2,3], wearable electronic [4,5], electronic skin [6–8], treatment [9,10], as shown in Fig. 1. However, in order to practically adapt to these emerging applications, flexible pressure sensors face challenges in terms of high sensitivity [11,12], high resolution [13], fast response [14,15], good stability [16], and strong robustness [17,18]. In addition, the flexible pressure sensor must be equipped with multi-function, self-powered, self-healing and other functions to realize the feasibility of application in various complex environments.

To date, much scientific research has focused on the development of flexible pressure sensors [19,20]. After decades of development, pressure sensors from traditional rigid sensors to flexible wearable sensors, not only the sensitivity of the pressure sensor has been greatly improved, but also endowed with excellent properties such as self-power supply, self-healing, multi-function and so on. With the exploration of flexible and functional materials, various conversion principles are widely used to prepare flexible pressure sensors. Many studies have also developed flexible pressure sensors by designing different microstructures [21,22,23]. In addition, in order to meet the requirements of industrial production and practical applications, many reports have focused on improving the sensitivity of flexible pressure sensors.

According to the transduction mechanisms, flexible pressure sensors are generally divided into three types: piezoresistive, capacitive and piezoelectric, as shown in Fig. 2. Based on the above three conversion principles, lots of work have been done to improve the properties of the flexible pressure sensor. The advantages and disadvantages of the three types of pressure sensors are compared as shown in Table 1. It can be seen that each sensor has its advantages and disadvantages, so we need to choose the matching sensor according to the actual application field. Piezoresistive pressure sensors can be applied in medical examination, sealing inspection, physical exercise, and so on. As we know, there are some commercial products in the market, such as the incredible smart bra, which can monitor the heartbeat by resistance variation to prevent the occurrence of diseases [96]. Capacitive pressure sensors can be applied in wearable respiratory monitoring, health monitor-

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Fig. 1. Applications of recently developed devices for flexible pressure sensors. “health monitor”, reproduced with permission [[12], [35]] [64],. Copyright 2017, American Chemical Society [12]; Copyright 2019, Elsevier [35]; Copyright 2020, Elsevier [64]. “e-skin”, reproduced with permission [26,88,91]. Copyright 2017, Wiley-VCH [26]; Copyright 2015, Springer Nature [88]; Copyright 2012, Springer Nature [91]. “wearable electronic”, reproduced with permission [8,54,55]. Copyright 2017, Wiley-VCH [8]; Copyright 2018, The Royal Society of Chemistry [54]; Copyright 2019, American Chemical Society [55]. “human-machine interface”, reproduced with permission [27,58,62]. Copyright 2019, American Chemical Society [27]; Copyright 2018, Springer Nature [58]; Copyright 2017, Wiley-VCH [62].

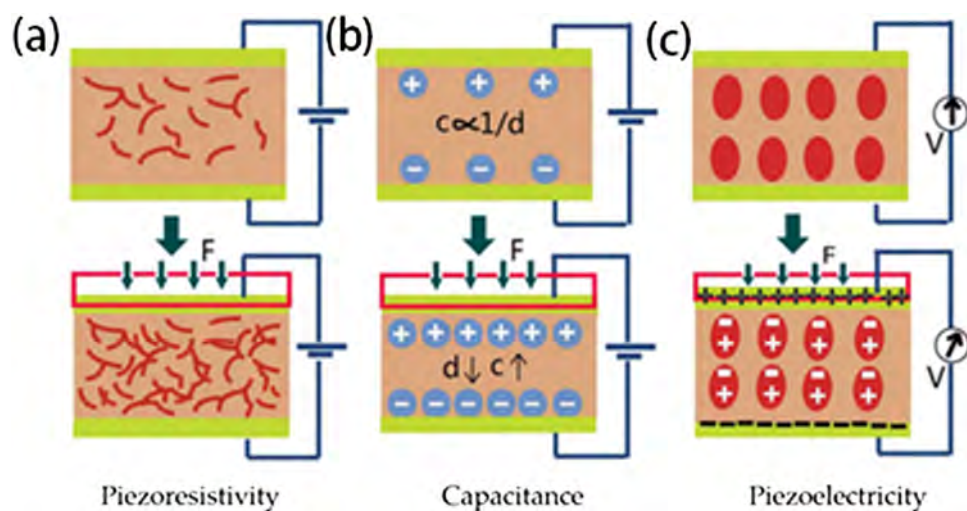


Fig. 2. The schematic images of transduction methods: (a) piezoresistivity, (b) capacitance, and (c) piezoelectricity.

Table 1

The advantages and disadvantages of the three kinds of pressure sensors.

Transduction principles	Advantages	Disadvantages
Capacitance	High sensitivity Simple structure Large dynamic range Fast response Suitable for small force test Good temperature stability	Poor linearity Affected by parasitic capacitance Susceptible to electromagnetic interference
Piezoresistivity	The manufacturing process is simple Low cost Strong anti-interference ability Easy to achieve small size Large deformation	Low sensitivity Poor stability Poor temperature stability
Piezoelectricity	High sensitivity Fast response Low power consumption	Low spatial resolution Only applicable to dynamic testing Poor stretchability

ing, human-computer interface, etc. Because of the characteristics of the capacitive pressure sensor, it can not only be applied to the temperature insensitive environment [50], but also can realize wireless transmission through the induction capacitor technology [48]. Piezoelectric pressure sensors have been widely used in wearable electronic devices, electronic skin and medical detection due to their self-powered characteristics.

At present, flexible pressure sensors still have many technical limitations and challenges [24,25,26,27,28]. First, flexible pressure sensors require higher sensitivity and wider detection range. With the advancement of science and the development of artificial intelligence, especially the arrival of 5 G, pressure sensors will be used more in ultra-precise fields, such as ultra-small pressure detection. This requires that the pressure sensor should have ultra-high sensitivity and a wide detection range at the same time. Second, the flexible pressure sensor requires low power consumption and multifunction. Low power consumption is the goal pursued by all devices, so flexible pressure sensors also need to pursue low power consumption, even self-powered. At present, most pressure sensors can only detect pressure alone, which is not conducive to practical applications. Designing a multi-functional flexible pressure sensor is also one of the challenges in the development of pressure sensors. Third, the traditional manufacturing methods of flexible pressure sensors (such as lithography, magnetron sputtering, etc.) are too complicated, with long manufacturing cycle and high cost, and a large amount of material waste will be generated in the manufacturing process. Therefore, a flexible pressure sensor method which can realize rapid manufacturing and save cost and resources is very important.

In this review, we highlight the basic design and performance improvements of flexible pressure sensors. In the second, third and fourth section, we introduced the latest research progress of piezoresistive flexible pressure sensors, capacitive flexible pressure sensors and piezoelectric flexible pressure sensors. In the fifth section, we discussed the design of high-performance flexible pressure sensors for different application requirements. For example, self-powered flexible pressure sensors, multifunctional flexible pressure sensors and self-healing flexible pressure sensors. Finally, we summarize the latest breakthroughs, new research directions, and new application directions for flexible pressure sensors.

2. Recent developments of piezoresistive pressure sensor

The conversion principle of the piezoresistive sensor is to deform the device by applying external pressure, which indirectly changes the distribution density and contact state of the conductive filler inside the sensor, and then causes the resistance of the sensor to change regularly. Therefore, they do not require complicated sensor structures, and compared with capacitive and piezoelectric

pressure sensors, their wide pressure test range, and simple manufacturing process have made such sensors widely studied. In order to improve the performance of piezoresistive pressure sensors, researchers usually use two methods: optimize the active material and improve the structure of the device. As shown in the following sections.

2.1. Piezoresistive pressure sensors with different materials

In the past decade, various materials have been selected to achieve the excellent performance of pressure sensors. Some researchers have demonstrated that carbon-based materials including carbon black, carbon nanotubes (CNT) [29–31], graphene [32–35] and polymers combined with metal nanowires [36,37], metal nanoparticles [38,39], etc. have significant piezoresistive properties.

Carbon nanotube was an excellent carbon-based material with excellent conductivity and flexibility, and was one of the excellent materials for making piezoresistive pressure sensors. In order to independently detect omnidirectional bending and pressure, Chen et al. used carbon nanotube-polyurethane (CNT-PU) sponge to develop a lightweight, low-cost, and retractable multifunctional sensor [29]. Fig. 3a shown the schematic illustration of the fabrication process of this flexible pressure sensor and Fig. 3b, c, d, e shown the sensor's ability to detect omnidirectional bending pressure. In 2017, Zhan et al. [30] made a flexible, wearable pressure sensor used single wall carbon nanotube (SWNTs) into tissue paper. The sensor exhibited excellent performance and had many advantages, including high sensitivity of 2.2 kPa^{-1} in the pressure range of 35–2500 Pa, and high sensitivity of 1.3 kPa^{-1} in the pressure range of 2500–11700 Pa, Ultra-low energy consumption ($\sim 10^{-6} \text{ W}$), and a rapid response time. The SWNT-based pressure sensor capable of detecting with high sensitivity to the soft touch of the human body, so that they can be ultra-low power (low power consumption $\sim \mu\text{W}$) to sense a number of important physiological signals of the human body. The pressure sensor can also be integrated into the soft skin, which can locate the location and even the amount of pressure applied to the robot. In the same year, Park et al. [31] used a shrinking manufacturing process to connect highly wrinkled CNT film electrodes face to face to produce a piezoresistive pressure sensor. Wrinkles can not only provide strain relief, but also increase pressure sensitivity by 12800 times, with a response time of less than 20 milliseconds. The improved sensitivity is attributed to the surface roughness of wrinkles. When two corrugated electrodes are coupled together, the number of electrical contacts changes upon actuation, thereby changing the resistivity.

Graphene, an atomically thin carbon material with remarkable conductivity, is one of the most widely investigated materials in

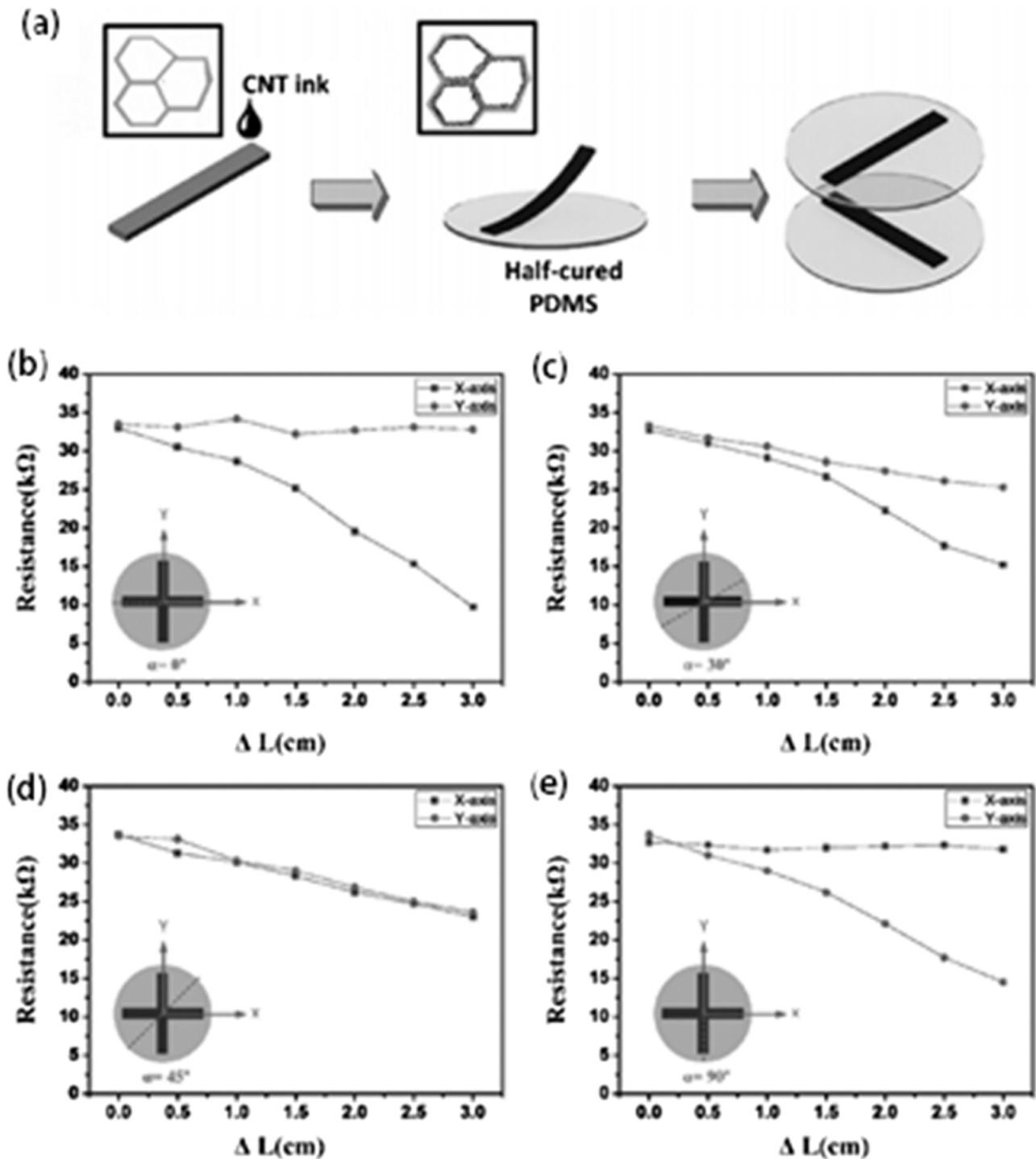


Fig. 3. (a) Fabrication process of the sensor, including dip CNT ink on the sponge, putting the CNT-PU sponge strips on the half-cured PDMS and assembling two functional layers perpendicularly and experimental result of the whole sensor bending at b) 0°, c) 30°, d) 45°, and e) 90°. Reproduced with permission [29]. Copyright 2017, Wiley-VCH.

the past few years. Since recent reviews on graphene have provided details thoroughly on its properties, we discuss only a few examples of its applications in pressure sensors. Rinaldi et al. [32] used PDMS foam coated with graphene nanosheets to prepare a flexible and highly sensitive pressure sensor. This sensor was loaded with 0.96 wt.% of multilayer graphene nanosheets. It was characterized by highly repeatable pressure-dependent conductance after several stable cycles. It was suitable for detecting compressive stress as low as 10 kPa, with a sensitivity of 0.23 kPa^{-1} , corresponding to an applied pressure of 70 kPa. In addition, it is estimated that the

sensor can detect pressure changes of $\sim 1 \text{ Pa}$. Tao et al. [33] mixed tissue paper with graphene oxide (GO) solution to obtain GO paper, thus fabricating a high-performance paper-based pressure sensor. Paper-based pressure sensors could measure wrist pulse, speech, breathing and movement status. The pressure range of the pressure sensor is 0–20 kPa, and the sensitivity was up to 17.2 kPa^{-1} , as shown in Fig. 4. Compared with the previously studied pressure sensors, achieved ultra-high sensitivity within a sufficient working range had obvious advantages, and could better meet the requirements of human physiological activity detection. Lou et al. [34]

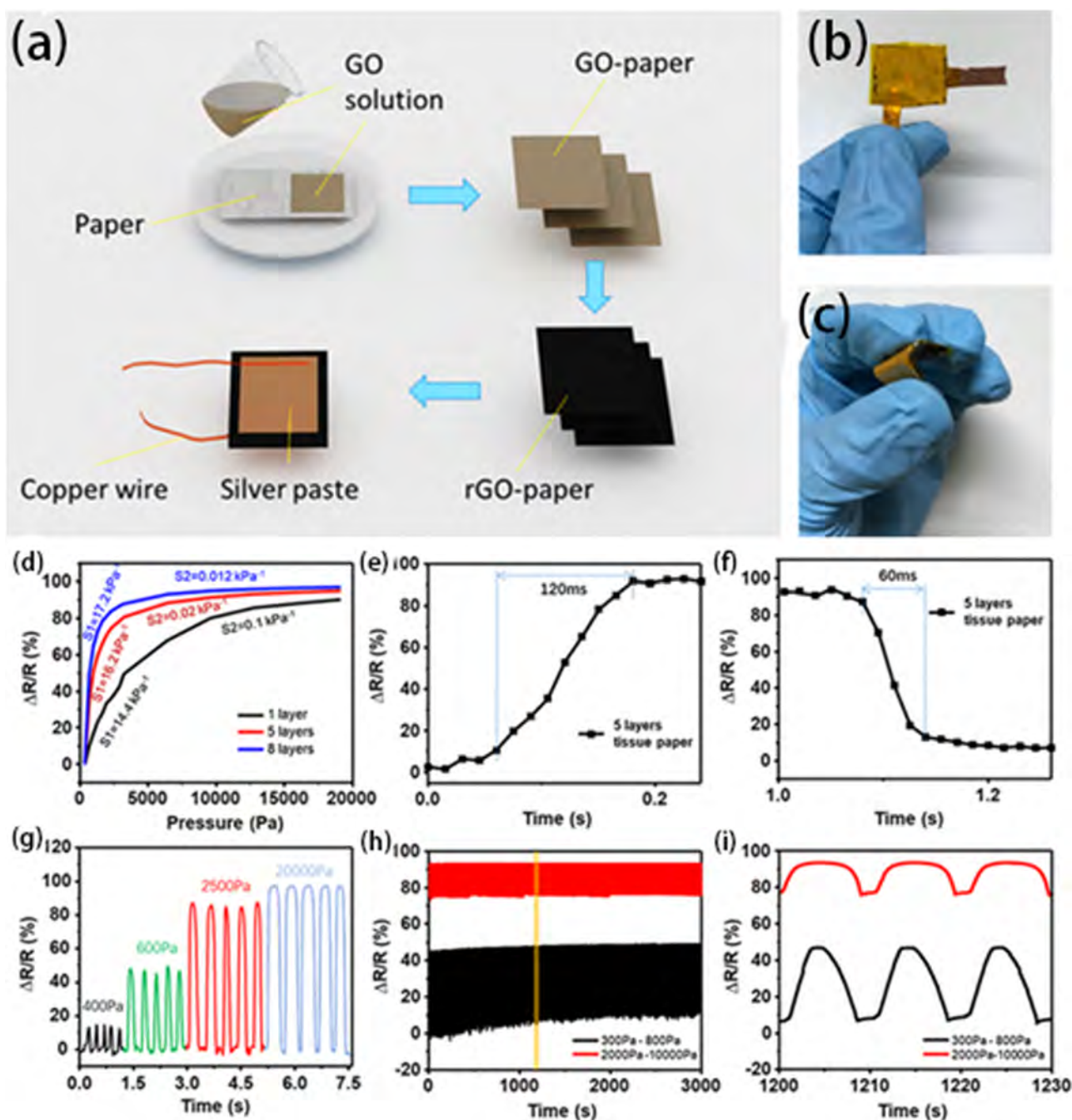


Fig. 4. (a) Process of the graphene-based pressure sensors with paper substrate. (b) Photo of pressure sensor encapsulated with PI. (c) Bent sensor showing good flexibility. (d) Change of resistance with pressure increases for graphene-based sensor with one, five, and eight layers of tissue paper. (e) Response time of tissue paper pressure sensor. (f) Recovery time of tissue paper pressure sensor. (g) Response test of graphene pressure sensor at different pressure. (h) Test of repeatability characteristics of 300 cycles. (i) Enlarged image of (h). Reproduced with permission [33]. Copyright 2017, American Chemical Society.

made a flexible pressure sensor by attaching conductive graphene films on the upper and lower parts of the fabric. The photograph of graphene textile was shown as inset in Fig. 5, which exhibited excellent flexibility. The sensitivity of the device could reach 0.012 kPa^{-1} , the measurement range could reach 800 kPa, and the response time could reach 50 ms. In 2019, He et al. [35] developed a high-performance piezoresistive pressure sensor based on high conductivity interface self-assembled graphene (ISG) film. The sensor had unprecedented comprehensive performance, with high sensitivity of 1875.53 kPa^{-1} and a wide linear detection range of 0–40 kPa. The sensor also had good stability and a high peak signal-to-noise ratio of 78 dB. It also had a high durability of 15,000 cycles,

a fast response of 0.5 ms and a recovery time of 0.8 ms, and a low detection limit of 1.8 Pa, as show in Fig. 6.

Carbon-based nanomaterials have broad application prospects in piezoresistive pressure sensors. However, the electrical conductivity of these carbon nanomaterials cannot reach its expected value because the conjugated structure may be destroyed during its manufacturing process (liquid exfoliation or redox process). Moreover, since more harmful solvents (concentrated H_2SO_4 , N_2H_4 , NMP, etc.) may be used, these carbon nanomaterials are expensive and environmentally unfriendly, which will greatly hinder their application. In recent years, metal nanowires and metal nanoparticles have attracted much attention and widely developed

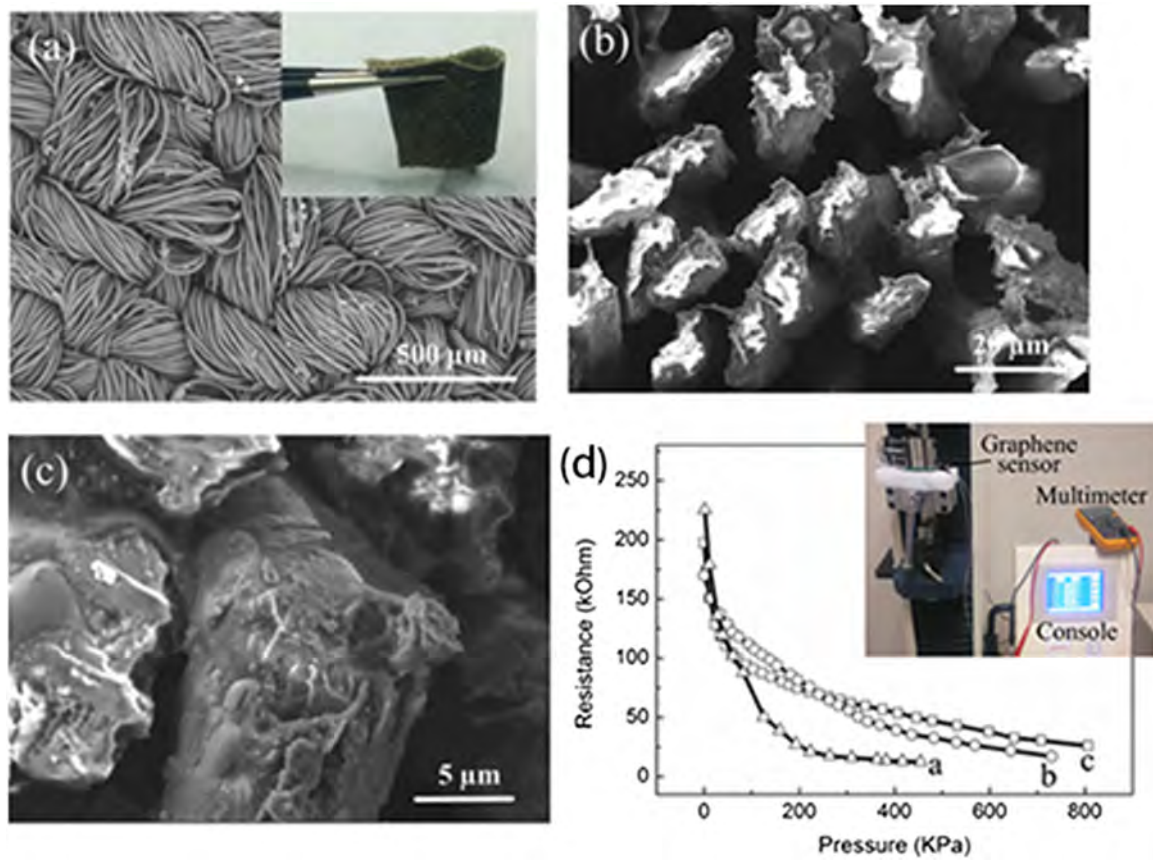


Fig. 5. (a) The photograph of graphene textile sensor under low magnification. (b) The cross-sectional SEM image of graphene fibres at 3000 $\times$ magnification. (c) at 10,000 $\times$ magnification. (d) The resistance curves of the graphene textile pressure sensors vs. applied loading. a: 2-layers textile sensor; b: 3-layers textile sensor; c: 4-layers textile sensor. Reproduced with permission [34]. Copyright 2017, MDPI.

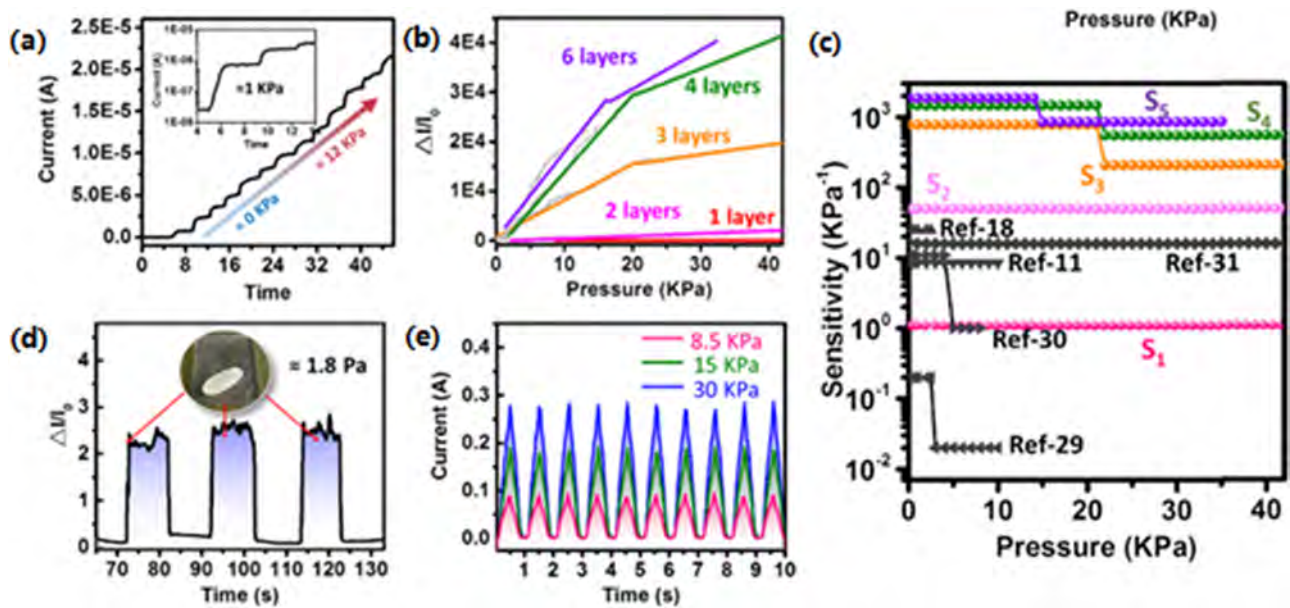


Fig. 6. (a) The current output of a pressure sensor constructed with two ISG film. (b) Pressure response sensitivity curves of pressure sensors constructed with different ISG membrane layers. (c) Compare with the performance of the previously reported piezoresistive sensor. (d) The four-layer ISG film sensor can detect very small pressures. (e) The real-time I_t curve of the four-layer ISG film sensor pressure sensor device responds to three large repeated applied pressures. Reproduced with permission [35]. Copyright 2019, Elsevier.

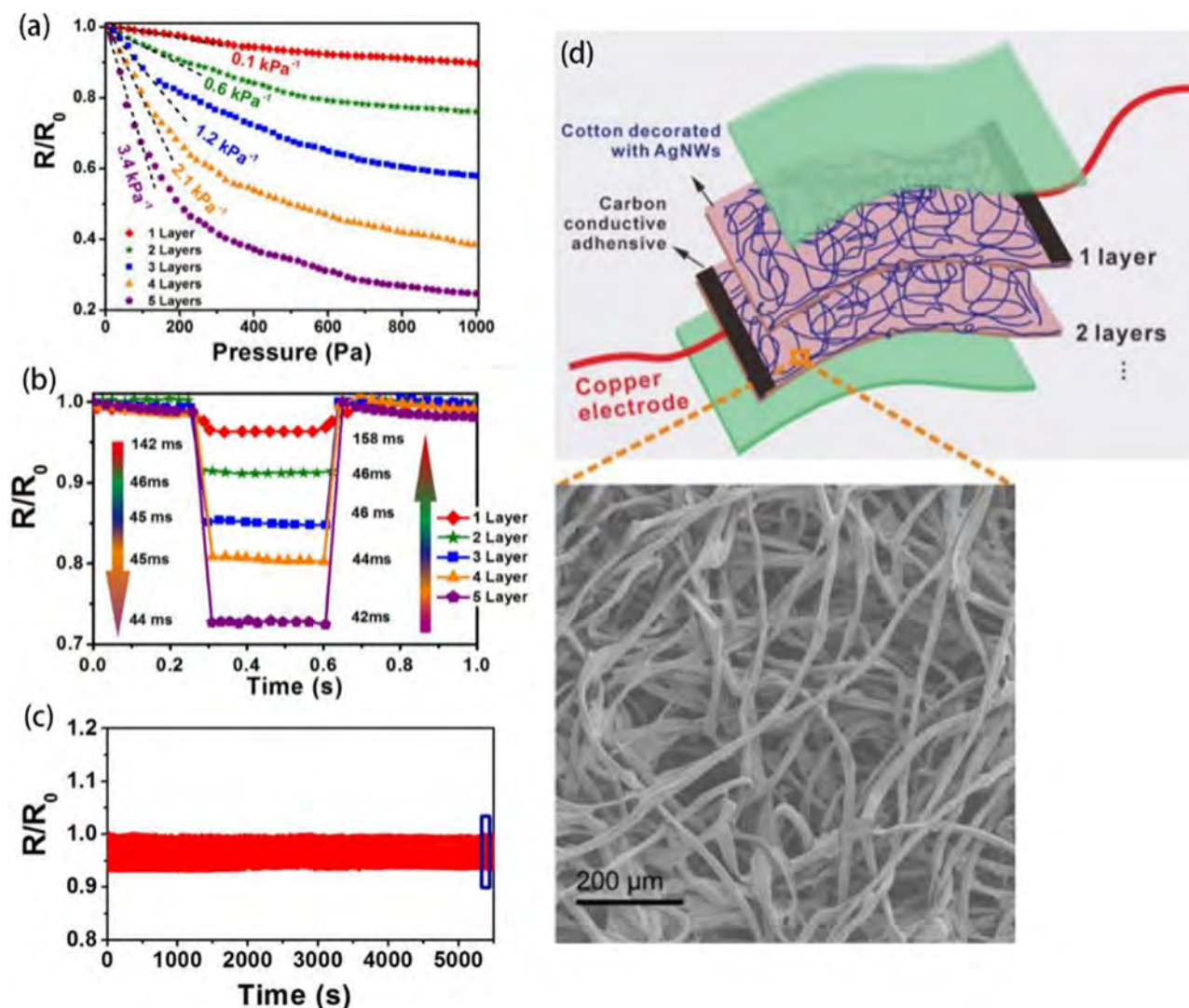


Fig. 7. (a) relative resistance change–pressure curve for the pressure sensor constructed by different layers of cotton sheets; (b) fast response and relaxation time for the flexible pressure sensors constructed by different layers of conductive cotton sheets; (c) normalized sensitivity of pressure sensors as a function of loading cycles (applied pressure is 100 Pa); (d) Schematic of the flexible pressure sensors. Reproduced with permission [36]. Copyright 2015, The Royal Society of Chemistry.

to construct the conductive networks for flexible piezoresistive materials because of their high conductivity and flexibility, easy synthesis and excellent anti-oxidation property, showing their potential applications in the field of flexible piezoresistive materials [36–39].

Wei et al. [36] manufactured flexible pressure sensors by coating silver nanowires on cotton. The flexible pressure sensors respond to the applied pressure from 0–1000 Pa is shown in Fig. 7a. As can be seen, a dramatic decreased in resistance was observed when the pressure applied on the pressure sensors and the layers of conductive cotton sheets exhibited a great influence on the sensitivity of pressure sensors. The sensitivity of pressure sensor with 3, 4 and 5 layers of cotton sheet were 1.2 kPa^{-1} , 2.1 kPa^{-1} and 3.4 kPa^{-1} . The sensor also had a fast response and relaxation time (<50 ms) and excellent stability (> 5000 load / unload cycles), as shown in Fig. 7b, c. A flexible piezoresistive pressure sensing matrix of copper nanowire composite aerogel was reported by Yap et al. [37] The sensors exhibited excellent sensitivity (6 kPa^{-1}) and durability (10,000 cycles cyclic) and could be easily scalable to form large-area sensing matrix for pressure mapping.

In 2016, Lee and his team reported by the highly sensitive one kind of sea urchin metallic nanoparticles (SSNP) and an insulat-

ing polyurethane (PU) elastomers composed of a transparent and durable piezoresistive pressure sensor [38]. Due to the effective quantum tunneling effect between SSNPs in the PU elastomer, it shown excellent pressure sensing performance with a high sensitivity of 2.46 kPa^{-1} . It also had excellent optical transmittance and mechanical flexibility. In addition, as shown in Fig. 8, it exhibited a fast response/relaxation time of 30 ms, excellent repeatability under pressure pulses of up to 600 s, and excellent operation durability of 200 bending cycles. Two years later, Lee and his team made improvements based on previous work [39]. They showed an ultra-sensitive and transparent piezoresistive pressure sensor based on a piezoresistive SSNPs-PU composite membrane with a microsphere array. The new piezoresistive material SSNP had an effective quantum tunneling effect even at very small concentrations, so it had excellent optical transmittance (77.7 % at 550 nm). The piezoresistive pressure sensor had an excellent sensitivity of 71.37 kPa^{-1} , which was due to the enhancement of the quantum tunneling effect caused by the stress concentration at the small contact point and the deformation of the contact area. It also had a fast response / relaxation time (30 ms), the ability to detect ultra-low pressure (4 Pa) and excellent long-term stability, as shown in Fig. 9.

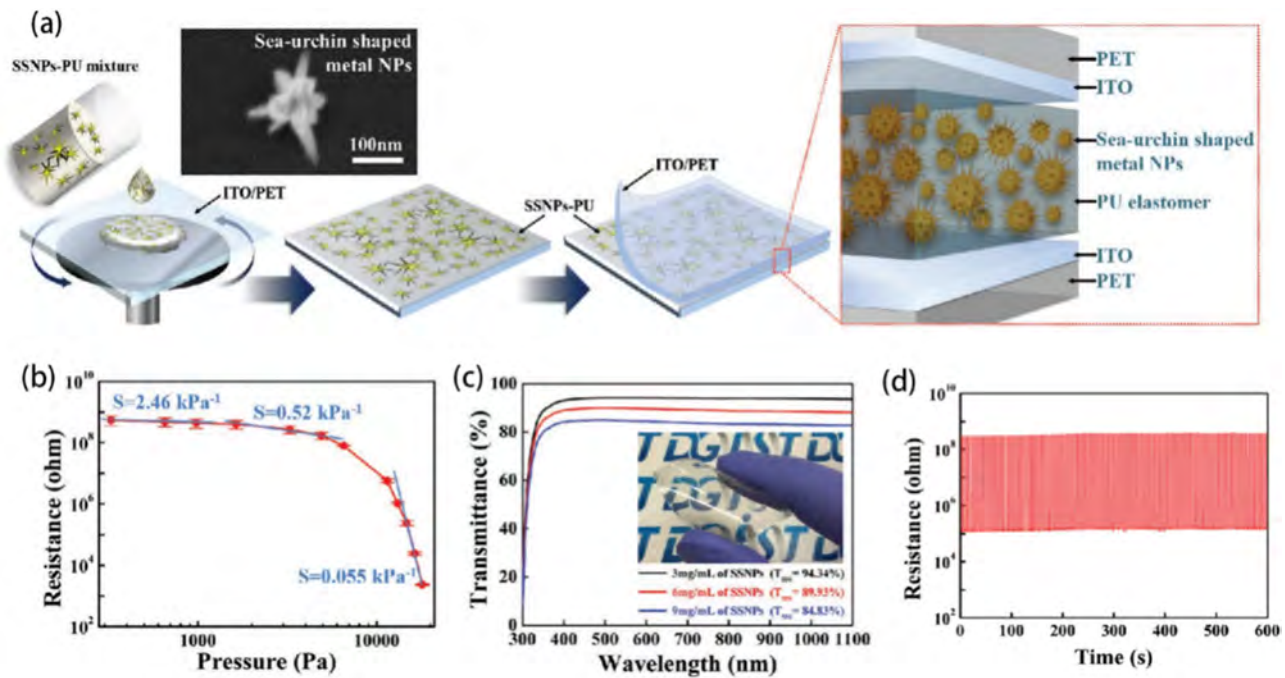


Fig. 8. (a) Schematic illustration for fabrication of a pressure sensor composed of SSNPs and PU. (b) Sensitivity of the pressure sensor under a wide range of applied pressures. (c) Optical transmittance of pressure sensors with various concentrations of SSNPs. Inset: A photograph of a pressure sensor. (d) Repeatability test under the pressure pulse of 14.7 kPa up to 600 s. Reproduced with permission [38]. Copyright 2016, Wiley-VCH.

2.2. Piezoresistive pressure sensors with different structures

In addition to using excellent materials to improve the piezoresistive pressure sensor, optimizing the structure of the piezoresistive pressure sensor is also an important method. Piezoresistive pressure sensors are usually constructed by coupling two conductive rough surfaces together. In this way, the number of electrical contacts can be changed by the applied mechanical pressure, thereby effectively increasing or decreasing the resistivity between the electrodes. In order to improve the performance of piezoresistive pressure sensors, researchers generally use two electrodes with rough surfaces, conductive porous sponge or foam, silk, paper-based and other methods to improve the performance of pressure sensors.

A flexible pressure sensor based on rGO/polyaniline wrapped sponge (RGPS) with tunable sensitivity was manufactured by Ge et al. [40], as shown in Fig. 10. From the flexible sensor, tunable sensitivity (0.042 to 0.152 kPa^{-1}), wide working range (0 – 27 kPa), fast response ($\sim 96 \text{ ms}$), high current output ($\sim 300 \mu\text{A}$ under 1 V), frequency-dependent performance, reliable repeatability (~ 9000 cycle) and stable signal waveform output could be readily obtained.

The performance of piezoresistive pressure sensors can also be improved by manufacturing irregular surface shapes of active materials [41–43]. The method of embedding vertically aligned carbon nanotube (VACNT) forests in PDMS with irregular surface morphology to obtain a highly compliant and robust carbon composite conductor was reported by Kim et al. [41]. The manufacturing and structure diagram of the sensor was shown in Fig. 11a. After assembled two rough composite conductors, the sensor shows excellent performance. The sensitivity was about 0.3 kPa^{-1} at pressures up to 0.7 kPa , and it had a stable steady-state response under various pressures, a wide detectable range of up to 5 kPa before saturation, a relatively fast response time of $\sim 162 \text{ ms}$, and good reproducibility over 5000 cycles of pressure loading/unloading, as shown in Fig. 12a. In addition to using the wrinkle of the substrate material, adding nanowires between the active material and the electrode is also a good way to produce irregular surfaces.

Liu et al. [42] developed a new pressure sensor by sandwiching internally connected polyvinyl alcohol (PVA) nanowires between a sheet of ultra-thin wrinkled graphene film (WGF) and a pair of interdigital electrodes (IDE). The IDE acted as soft substrate and provided conductive path. The manufacturing and structure diagram of the sensor was shown in Fig. 11b. The pressure sensor showed a high sensitivity of 28.34 kPa^{-1} . In addition, the device could recognize about 22.4 mg ($\sim 2.24 \text{ Pa}$) of lightweight rice, and had shown excellent durability and reliability after 6000 repeated loading and unloading cycles, as shown in Fig. 12b.

In order to improve the sensitivity of piezoresistive pressure sensors, a lot of research has been done. However, piezoresistive pressure sensors still have many challenges. First, the piezoresistive pressure sensor has great hysteresis, which limits the application of the device. Second, the temperature stability of the piezoresistive pressure sensor is very poor, which makes it difficult to work in an environment with large temperature changes.

3. Recent developments of capacitive pressure sensor

Capacitive flexible pressure sensor is based on the principle of parallel plate capacitors. In commonly used parallel plate capacitors, the capacitance is given by the formula $C = \epsilon_r \epsilon_0 A / d$, where C is the capacitance, ϵ_r is the relative electrostatic permittivity of the material between the plates, ϵ_0 is the electrical constant, and A is the area where the two plates overlap Square meters, d is the spacing between the plates (in meters). The equation has three variables (ϵ_r , A and d), and ϵ_r is always constant, while d and A are easily changed by external forces. The applied pressure causes the plates to deform and the capacitance to change. Capacitive pressure sensors have been extensively studied due to their large dynamic range, high sensitivity, fast response, simple structure and suitable for small force testing. In order to improve the performance of capacitive pressure sensors, researchers generally focus on improving the materials and structures of the dielectric layer and electrodes.

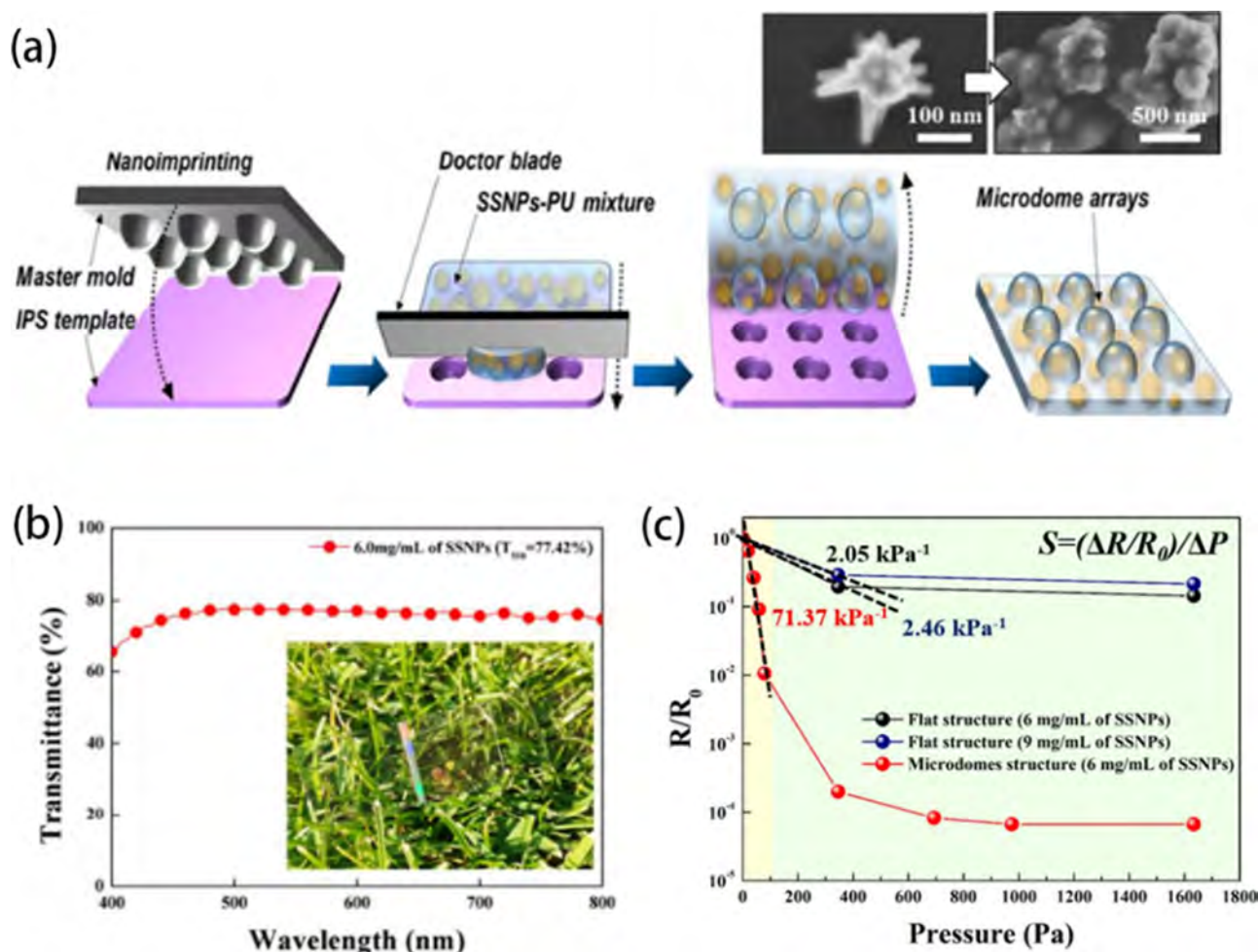


Fig. 9. (a) Schematic illustration for the fabrication of a SSNPs-PU composite film with microdomes. Inset: SEM image of a SSNP and SSNP-PU mixture (b) Optical transmittance of microdome structured pressure sensors with different concentrations of SSNPs (6.0 mg/mL). Inset: A photograph of a piezoresistive pressure sensor over artificial turf. (c) Sensitivity of the pressure sensor under a wide range of applied pressures. Reproduced with permission [39]. Copyright 2018, The Royal Society of Chemistry.

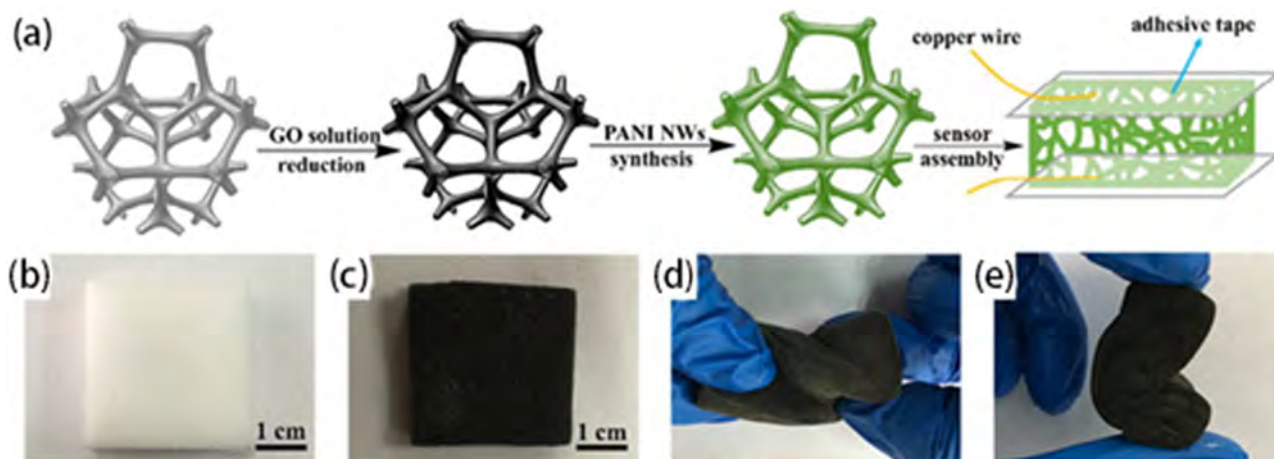


Fig. 10. (a) Fabrication process of the flexible pressure sensor. (b, c) Photographs of pristine sponge and RGPS. (d, e) Photographs of the twisted and folded RGPS. Reproduced with permission [40]. Copyright 2018, The Royal Society of Chemistry.

3.1. Capacitive pressure sensor with different dielectric layer

For capacitive pressure sensors, the capacitance is determined by the relative dielectric constant (ϵ_r), area (A) and the distance between the electrodes (d). All these variables (ϵ_r , A and d) may

be sensitive to pressure changes [46,47]. When an external force is applied to the soft pressure sensor, the thickness of the dielectric layer of the capacitive sensor changes, which causes the capacitance of the sensor to change. However, the sensitivity of capacitive sensors with elastomer dielectrics is limited by the compressibil-

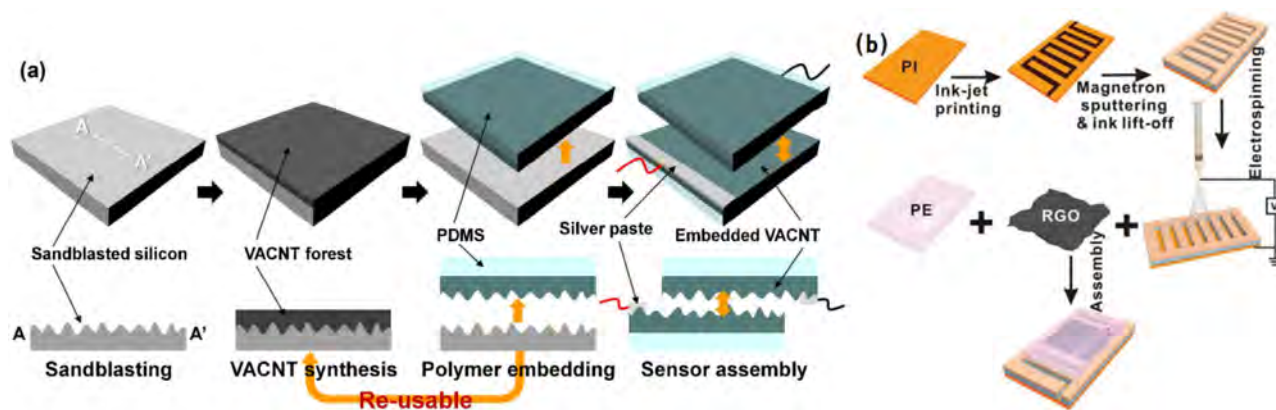


Fig. 11. (a) schematic illustration of the fabrication process of the flexible pressure sensor based on flexible VACNT/PDMS composite conductors with irregular surface morphology. Reproduced with permission [41]. Copyright 2017, American Chemical Society. (b) The schematic of the fabrication of piezoresistive pressure sensor based on synergistical innerconnect PVA nanowires/WGF. Reproduced with permission [42]. Copyright 2018, Wiley-VCH.

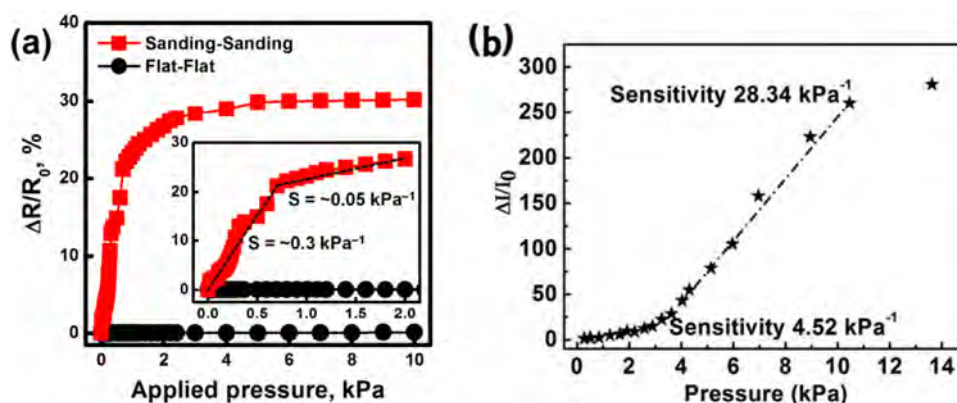


Fig. 12. (a) change in the electrical resistance of the flexible pressure sensor based on flexible VACNT/PDMS composite conductors (inset: magnified sensor responses representing pressure-sensitivities). Reproduced with permission [41]. Copyright 2017, American Chemical Society. (b) The relation between $\Delta I/I_0$ and ΔP on the fabrication of piezoresistive pressure sensor based on synergistical innerconnect PVA nanowires/WGF. Reproduced with permission [42]. Copyright 2018, Wiley-VCH.

ity of the elastomer, and the capacitance change of parallel plate sensors under load is relatively small, so the achievable sensitivity is usually very low. Therefore, many studies have focused on the modification of the dielectric layer to improve sensitivity.

The dielectric layer with a microporous structure can be highly deformed under a small pressure input level, thus greatly improving the sensitivity. At present, there are many ways to obtain microporous structures. Deionized water boiling point above the curing temperature characteristics of PDMS to manufacture a porous film was a good method. Lee and his co-worker [43] developed a low-cost flexible pressure sensor based on an elastomer film with uniformly distributed micropores as a dielectric layer. The porous film was fabricated simply using a mixture of an elastomer material of siloxane polymers and water droplets without any additive such as an emulsifying agent, as shown in Fig. 13a. The sensor shown a high sensitivity of 1.18 kPa^{-1} at low pressure ($< 0.02 \text{ kPa}$) and had a fast response time of 150 ms. Used a solid template to obtain a porous dielectric layer was also a common method. Kwon et al. [44] formed a porous Ecoflex dielectric layer by molding Ecoflex elastomer in a sugar cube template. The pressure sensor exhibited excellent performance with a high sensitivity of 0.601 kPa^{-1} in a low-pressure regime ($< 5 \text{ kPa}$) and a wide dynamic range of $0.1 \text{ Pa} - 130 \text{ kPa}$. Kang et al. [45] used polystyrene beads with a certain diameter to prepare the porous structure of PDMS dielectric layer, as shown in Fig. 13b. The sensor had a high sensitivity of 0.63 kPa^{-1} at low pressure, a response and relaxation time of about 40 ms, an ultra-low pressure detection of 2.42 Pa ,

and excellent durability and stability over 10000 working cycles. In addition to the above two methods, a dielectric layer with a microporous structure could also be obtained by generating gas in PDMS through a chemical reaction. Chen et al. [49] prepared a porous dielectric layer by added ammonium bicarbonate (NH_4HCO_3) to PDMS, which greatly improved the linear response range of the sensor. Kou et al. [48] added NH_4HCO_3 to PDMS and stir well, then heat to decompose NH_4HCO_3 to obtain PDMS sponge, as shown in Fig. 13c. This dielectric layer had high performance, high sensitivity, wide working range ($0 - 500 \text{ kPa}$), fast response time ($\sim 7 \text{ ms}$), low detection limit (5 Pa), and good stability, recoverability and repeatability. They sandwiched a sponge between the two surfaces of a folded flexible printed circuit, and used copper as an antenna and electrode to obtain a wireless pressure sensor.

The micropore structure can improve the sensitivity of the capacitive pressure sensor. However, introducing holes in the dielectric layer will inevitably lower the dielectric constant and make the original capacitance smaller, and eventually encounter the limitation of continuing to increase sensitivity. In order to improve the sensitivity and related performance of the capacitive sensor, the dielectric layer must have a more obvious deformation in the constant pressure range. In addition to the microporous structure dielectric layer, other microstructured dielectric layers was also effective methods. Pyramidal microstructure was one of effective structure [50–53]. Yang et al. [50] had manufactured a novel porous pyramid dielectric layer (PPDL) to achieved unprecedented sensitivity in capacitive pressure sensors. In the pressure

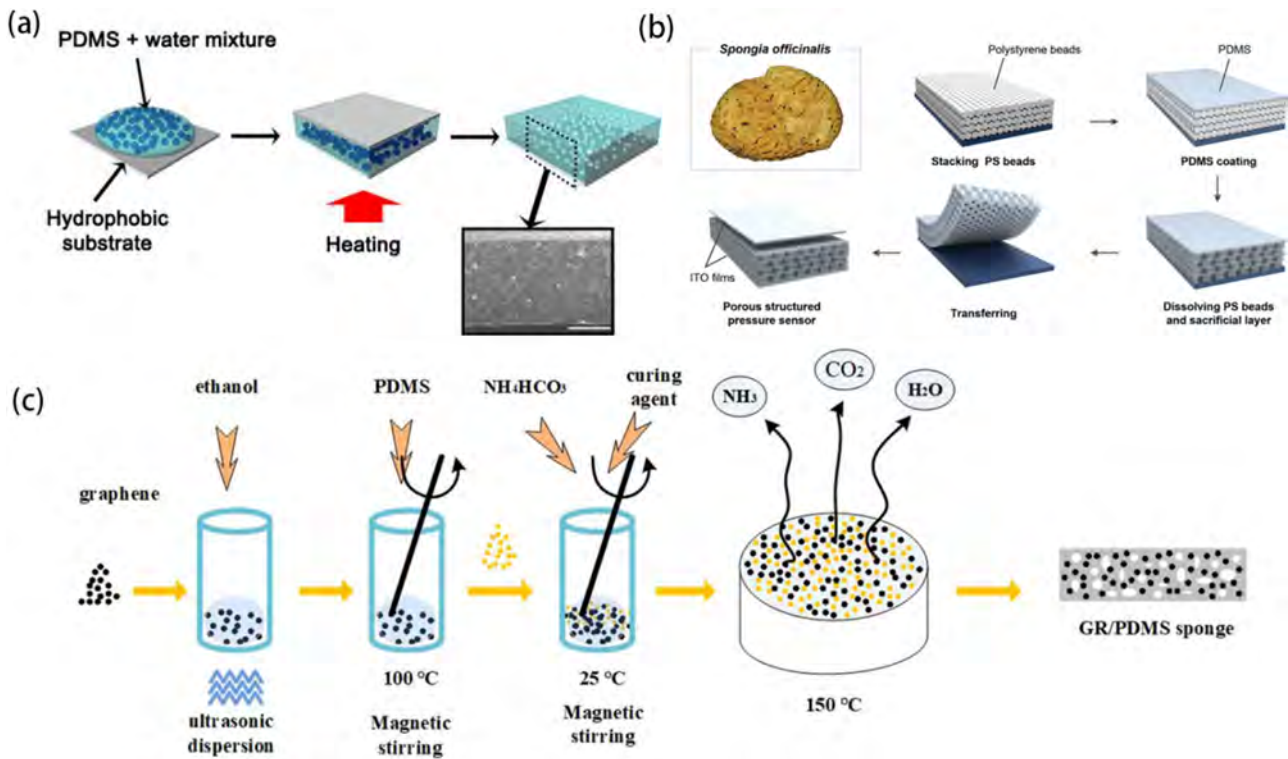


Fig. 13. Fabrication process of an elastomer film with well-distributed micro-pores. (a) Disperse DI water in PDMS to get microporous dielectric layer. Reproduced with permission [43]. Copyright 2016, Elsevier. (b) a porous structured PDMS film fabricated using PS beads. Reproduced with permission [45]. Copyright 2016, Wiley-VCH. (c) Decompose NH_4HCO_3 by heating to obtain PDMS sponge. Reproduced with permission [48]. Copyright 2019, Springer Nature.

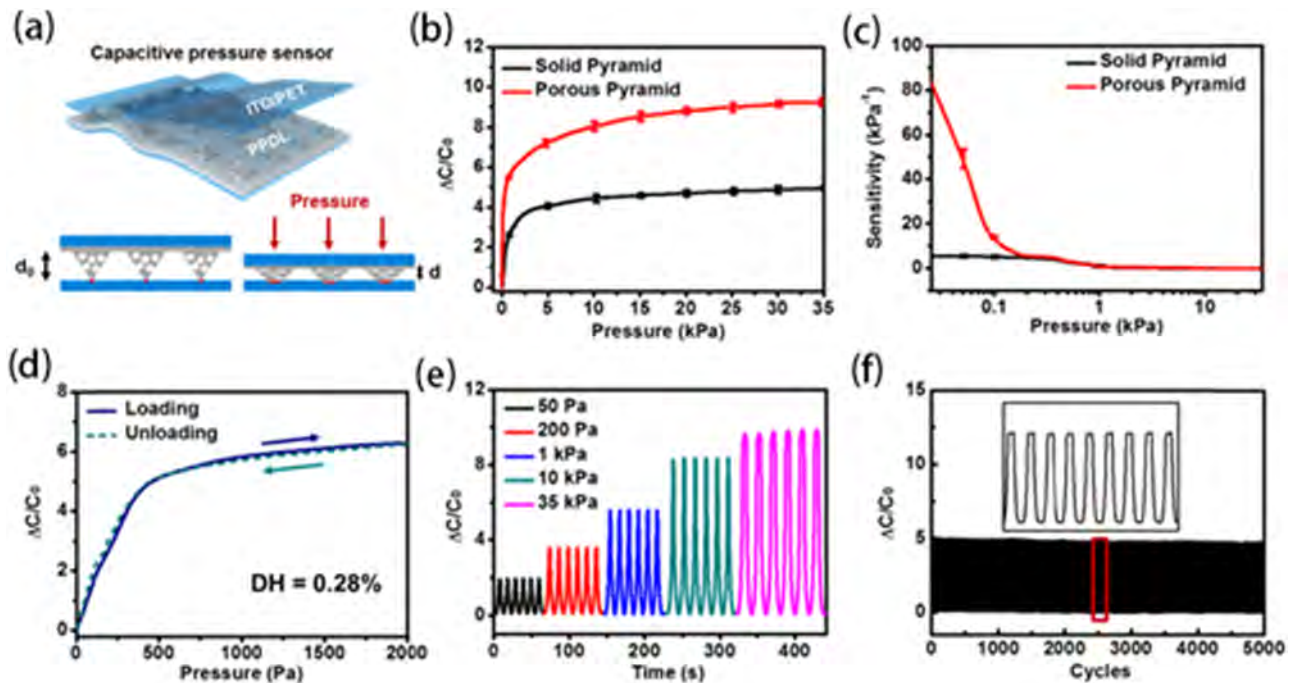


Fig. 14. (a) Schematic of capacitive pressure sensor based on PPDL. (b, c) Relative change in (b) capacitance and (c) sensitivity versus pressure of the capacitive pressure sensor based on solid pyramid dielectric layer (SPDL) and PPDL. (d) Hysteresis curve of the PPDL-based capacitive pressure sensor (strain rate: 1 mm/min). (e, f) Relative change in capacitance versus time plots of the PPDL-based capacitive pressure sensor under repeated pressure of (e) 0.05, 0.2, 1, 10, and 35 kPa and under (f) 5,000 cycles at 400 Pa. Reproduced with permission [50]. Copyright 2019, American Chemical Society.

range of <100 Pa, the sensitivity of the capacitive sensor was 44.5 kPa^{-1} , as shown in Fig. 14. The increased in sensitivity was due to a lower compression modulus and a larger change in effective dielectric constant under pressure. The sensor not only had ultra-

high sensitivity but also had no response to stress and temperature. Ruth et al. [51] and Bao [52] et al. further improved the sensitivity of the pressure sensor based on the pyramid microstructure by modulating the microstructure.

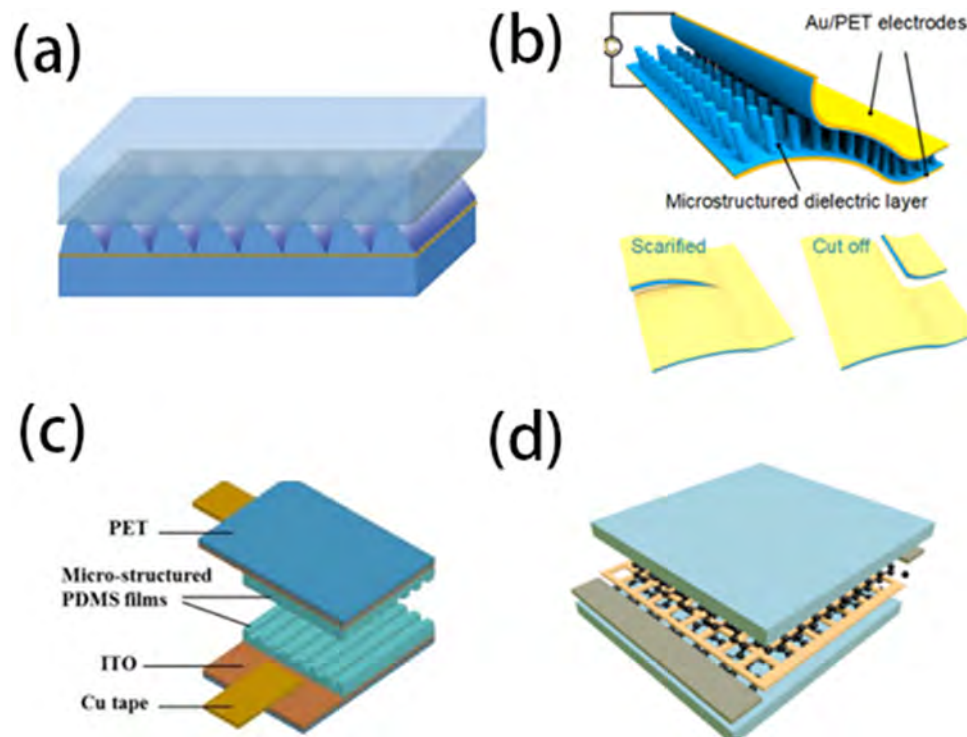


Fig. 15. different micro-structured dielectric layers of capacitive pressure sensor. (a) capacitive pressure sensor with a micro-array structured dielectric layer. Reproduced with permission [54]. Copyright 2018, The Royal Society of Chemistry. (b) capacitive sensor with a tilted micro-pillar array-structured dielectric layer. Reproduced with permission [55]. Copyright 2019, American Chemical Society. (c) The PDMS elastomer dielectric layer with microstructure in the capacitive flexible pressure sensor is obtained by 3-D printing manufacturing mold. Reproduced with permission [56]. Copyright 2017, IEEE. (d) a piezocapacitive pressure sensor based on the elastic nylon netting dielectric layer. Reproduced with permission [57]. Copyright 2018, American Chemical Society.

Table 2

Summarizes data on the sensitivity, response time, detection limit, and stability of capacitive pressure sensors with different dielectric layers.

The material of the dielectric layer.	The structure of the dielectric layer.	Sensitivity(kPa^{-1})	response time(ms)	limit of detection	Stability(cycles)	Ref
PDMS	microporous	1.18	150			[43]
Ecoflex	microporous	0.601		0.1		[44]
PDMS	microporous	0.63	~40	2.42	10000	[45]
PDMS/AgNWs	microporous	0.161			6000	[47]
graphene/PDMS	spong	2.2 MHz/kPa	~7	5	5000	[48]
PDMS	microporous	0.26	15	1	3000	[49]
PDMS	porous pyramid	44.5	50		5000	[50]
PDMS	microarray	2.04	100	7	1000	[54]
PDMS	Tilted micropillars	0.42		1	1000	[55]
PDMS	micro-structure	1.62			1000	[56]
nylon netting	nylon netting	0.33	20	3.3	1000	[57]
PDMS/AgNWs	micro-structure	0.831	30	1.4	10000	[58]

Other microstructured dielectric layers have been studied by many research groups. Ma et al. [54] reported a highly sensitive and flexible capacitive pressure sensor with a PDMS dielectric layer of a microarray structure via a simple strategy, as shown in Fig. 15a. The PDMS flexible substrate coated with silver nanowires (AgNWs) was used as the top/bottom electrode material, and a PDMS dielectric layer with a microarray structure was used to ensure high sensitivity of the pressure sensor. The sensors exhibited excellent performance, high sensitivity of 2.04 kPa^{-1} in low pressure ranges (0–2000 Pa), low detection limits ($<7 \text{ Pa}$) and fast response times ($<100 \text{ ms}$). The sensor also possessed high bending stability after a 1000 cycle bending test and high work stability for a multiple pressure cycle test. Bending deformation rather than compression deformation of the dielectric layer is also a good way to improve the sensitivity of capacitive pressure sensors. Luo et al. [55] developed a capacitive sensor reinforced by an inclined micropillar array structure dielectric layer, as shown in Fig. 15b. Since the slanted micropillar undergo bending deformation rather than compression

deformation, the distance between the electrodes was easier to change, resulted in high pressure sensitivity of 0.42 kPa^{-1} and very small detection limit of 1 Pa . In addition, the slanted micropillar array structure eliminated the air gap at the interface between the structured dielectric layer and the electrode, and the strongly bonded between the dielectric layer and the electrode bestowed the sensor high stability and reliability. Zhuo et al. [56] used 3-D printing technology to fabricate molds with a PDMS elastomer dielectric layer in a microstructured capacitive flexible pressure sensor, as shown in Fig. 15c. The device had a very low detection limit, short response/recovery time, excellent durability, and good tolerance to change in ambient temperature and humidity, enabling reliable real-time monitoring of weak human physiological signals. He et al. [57] proposed a piezoelectric capacitive pressure sensor based on an elastic nylon netting dielectric layer, as shown in Fig. 15d. The low-cost nylon netting dielectric layer with different thicknesses and mesh numbers insulated the two graphene electrodes, so the basic pressure sensor had an excellent

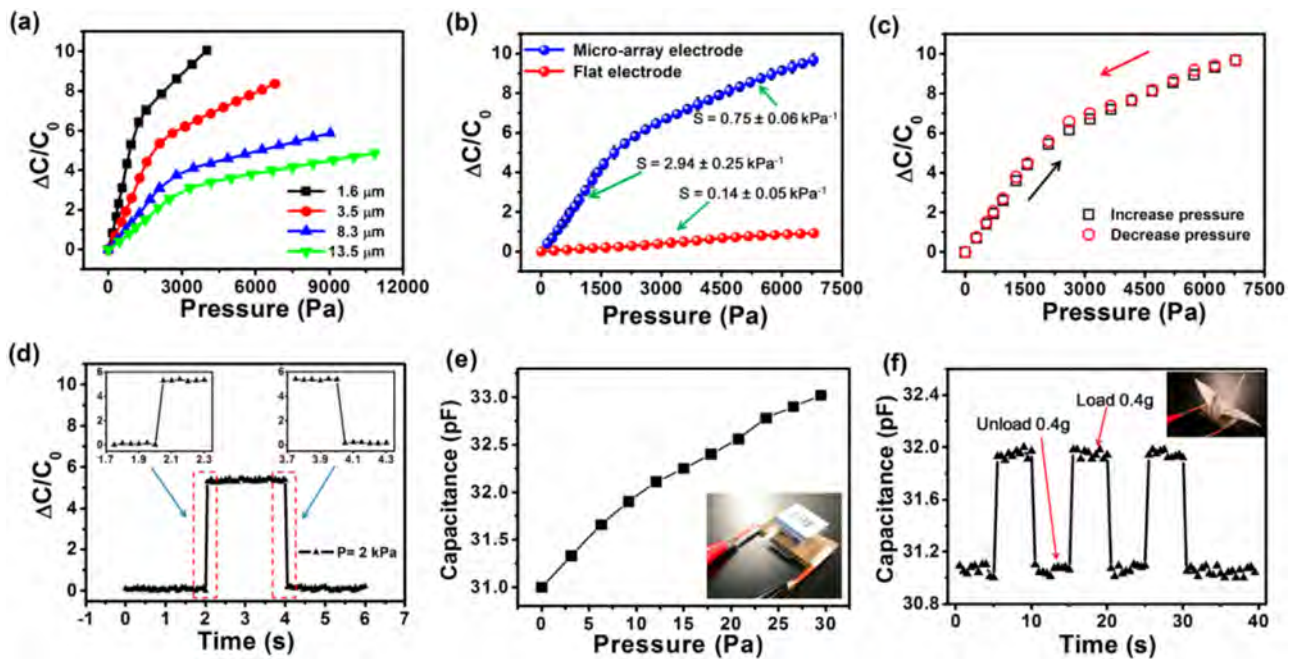


Fig. 16. (a) Thickness influence of PVDF dielectric layer on the performance of capacitive pressure sensor. (b) Relative capacitance variation – pressure curves of the capacitive pressure sensors based on PDMS microarray electrode and flat electrode. (c) Relative capacitance variation– pressure curves of the consecutive loading – unloading cycle. (d) Pressure response and relaxation time of the pressure sensor. (e) Pressure response of the capacitive pressure sensor at low pressure range. (f) Pressure response of the sensor for seasonal low pressure. Reproduced with permission [61]. Copyright 2017, American Chemical Society.

pressure sensing sensitivity of 0.33 kPa^{-1} at a pressure of 1 kPa, and had an ultra-low detection of 3.3 Pa after more than 1000 loading and unloading cycles, the limit had excellent mechanical stability. In addition, the sensor had an ultra-fast response speed of 20 ms, made it possible to detect small changes in pressure applied during changes in the shape of droplets that fall on the sensor. Shi et al. [58] used a simple and low-cost process to fabricate flexible, transparent and ultrasensitive capacitive pressure sensors by introducing patterned microstructured AgNW/PDMS composite dielectric films. The manufactured capacitive pressure sensor had high sensitivity (0.831 kPa^{-1}) when different pressures were applied, and had a detection limit of 1.4 Pa. In addition, capacitive pressure sensors had fast response (<30 milliseconds) and relaxation (<60 milliseconds) time. After more than 10,000 repeated cycles, it also exhibited high stability and durability. Table 2 summarizes some reported results of performances of flexible capacitive pressure sensor with different dielectric layer.

3.2. Capacitive pressure sensor with different electrodes

In addition to using excellent dielectric layers to improve capacitive pressure sensors, optimizing the electrodes of capacitive pressure sensors is also an important method. The modification of the dielectric layer can greatly improve the sensitivity of the capacitive pressure sensor. However, the sensitivity of capacitive sensors with elastomeric dielectrics is limited by the compressibility of the rubber. In order to further improve the performance of capacitive pressure sensors, researchers have also done a lot of research on the modification of electrodes.

In recent years, many high-sensitivity capacitive sensors with different electrodes have been reported. Cui et al. [59] demonstrated a flexible capacitive pressure sensor used to develop ultra-sensitive CNT/PDMS composite elastomer dielectric layer and Ag wrinkled electrode. The Ag wrinkled electrode was formed by vacuum deposition on a pre-strained and relaxed PDMS substrate, which was processed using O_2 plasma, surface functionalization

process and magnetron sputtering process. The developed sensor had a maximum sensitivity of 0.198 kPa^{-1} for capacitance, excellent durability in 500 cycles, and a fast mechanical response (<200 ms). Shuai et al. [61] also developed a very good pressure sensor using wrinkle electrodes. Unlike Cui et al., after plasma treatment with dry, low-pressure air, the prestretch PDMS film showed a spontaneous bending surface structure that was used to manufacture microarray molds. Using the mold, AgNW was transferred and embedded in a flexible microarray substrate and an elastic electrode was established. The capacitive pressure sensors possessed the superiorities of high sensitivity (2.94 kPa^{-1}), low detection limit (<3 Pa), short response time (<50 ms), excellent flexibility, and long-term cycle stability as shown in Fig. 16. Ataray et al. [60] used conductive fabric as electrode and organosilicon with microporous structure as dielectric layer to produce a highly sensitive and flexible pressure sensor for wearable electronic applications. Microstructure dielectric improved sensitivity, conductive fabric electrodes ensured a safe conductive network, and strong and flexible electrical connections were created through hot pressing and adhesive film technology. The combination of the conductive knit electrode with high porosity yielded the highest sensitivity of $121 \times 10^{-4} \text{ kPa}^{-1}$. Xiong et al. [64] used two PDMS-Au electrodes with surface convex microarrays and an ultra-thin PVDF dielectric layer to integrate a capacitive pressure sensor. The flexible capacitive pressure sensor exhibited an ultrahigh sensitivity of 30.2 kPa^{-1} (<130 Pa), low detection limit of 0.7 Pa, fast response time of 25 ms and excellent stability of 100,000 cycles. Yang and his co-worker [65], demonstrated a novel three-dimensional microconformal graphene electrode for ultrasensitive and tunable flexible capacitive pressure sensors. Microconformal graphene electrodes were obtained by transferring graphene onto poly ethylene terephthalate (PET) substrates by chemical vapor deposition. The flexible capacitive pressure sensor shown a high sensitivity of 3.19 kPa^{-1} , fast response of 30 ms and ultralow detection limit of 1 mg. Joo et al. [66] developed a capacitive pressure sensor based on a robust and elastic multi-scale structure PDMS

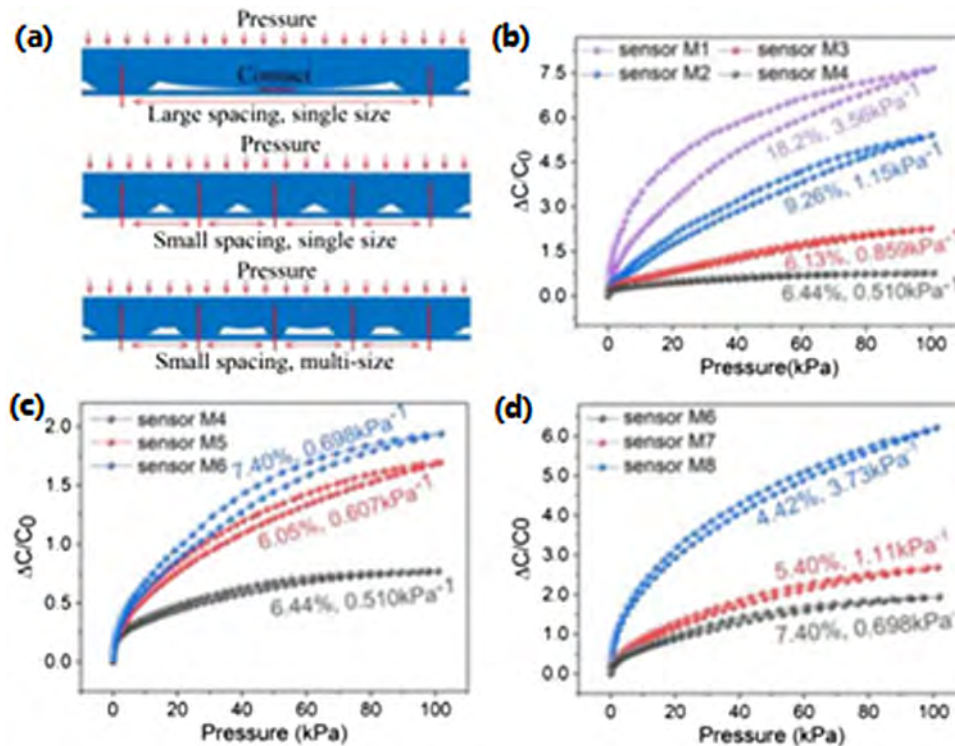


Fig. 17. (a) Schematic illustration how hierarchical microstructures reduce interfacial adhesion of electrodes. Piezocapacitance of sensors with: (b) pyramids of the same size but different spacing; (c) pyramids of different sizes but the same spacing, (d) pyramids of multi-size and the same spacing. Reproduced with permission [67]. Copyright 2017, IEEE.

Table 3

summarizes data on the sensitivity, response time, detection limit, and stability of capacitive pressure sensors with different electrodes.

The material of the electrodes	The structure of the electrodes	Sensitivity(kPa^{-1})	response time(ms)	limit of detection	Stability(cycles)	Ref
Ag	wrinkled	0.198	<200		500	[59]
conductive fabric	fabric	121×10^{-4}		0.86KPa	100	[60]
AgNWs	microarray	2.94	<50	3Pa		[61]
pencil	rough surface of paper	0.62		6Pa	5000	[62]
AgNWs	single layer				10000	[63]
Au	convex microarray	30.2	25	0.7Pa	100000	[64]
graphene	microconformal	7.68	30	1mg		[65]
AgNWs	multiscale	>3.8	<150		1500	[66]
Pt	hierarchically pyramid	3.73		0.1 Pa		[67]

electrode embedded with AgNWs. The sensor had high pressure sensitivity 3.8 kPa^{-1} ($45 \sim 500 \text{ Pa}$), and could detect very small pressure of 15 Pa . The sensor also shown fast response times of $< 150 \text{ ms}$, high stability for repeated cycles over 1500 times and high bending stability. Cheng et al. [67] reported the design of a flexible capacitive pressure sensor with a layered microstructure electrode, which can reduce the possible hysteresis caused by the interface and improve the sensitivity of the pressure sensor. The sparse large pyramid microstructure can improve the sensitivity, while the small pyramid microstructure can reduce the hysteresis caused by interface adhesion, as shown in Fig. 17. The optimized sensor had excellent performance, such as high sensitivity ($\sim 3.73 \text{ kPa}^{-1}$), ultra-low detection limit (0.1 Pa), significantly reduced hysteresis ($\sim 4.42\%$) and enhanced induction. Quan et al. [68] reported flexible capacitive micro-structured sensors based on a sandwich structure by laminating the silver nanowires (AgNWs) composite and the polydimethylsiloxane (PDMS) film together with PDMS as a dielectric layer. The sensor shown a high sensitivity of 1.1 kPa^{-1} , low detection limit of 1 Pa , a fast response time of $< 1 \text{ s}$ and high stability. Table 3 summarizes some reported results of performances of flexible capacitive pressure sensor with different dielectric electrodes.

Capacitive pressure sensors have been extensively studied, but they still have many difficulties to overcome. On the one hand, it is necessary to find better quality dielectric layer materials. For the dielectric layer, it is difficult to achieve high sensitivity of the sensor due to the high Young's modulus of the elastomer material. At present, most of the dielectric layer materials of capacitive pressure sensors are materials such as PDMS and Ecoflex. Although the Young's modulus of these materials is as low as 5 kPa , they still limit the increase in sensitivity of the pressure sensor. On the other hand, the detection range of capacitive pressure sensors needs to be further improved. Although most of the current capacitive pressure sensors exhibit good sensitivity and linearity at low pressures, their sensitivity and other performance at high pressures are not satisfactory.

4. Recent developments of piezoelectric pressure sensor

Many research groups have reported flexible pressure sensors based on various transduction mechanisms, including piezoresistive, capacitive, etc. However, these pressure sensors often require an external power source to drive them, which is one of the key obstacles for wearable/Internet of Things (IoT) sensors. Frequent

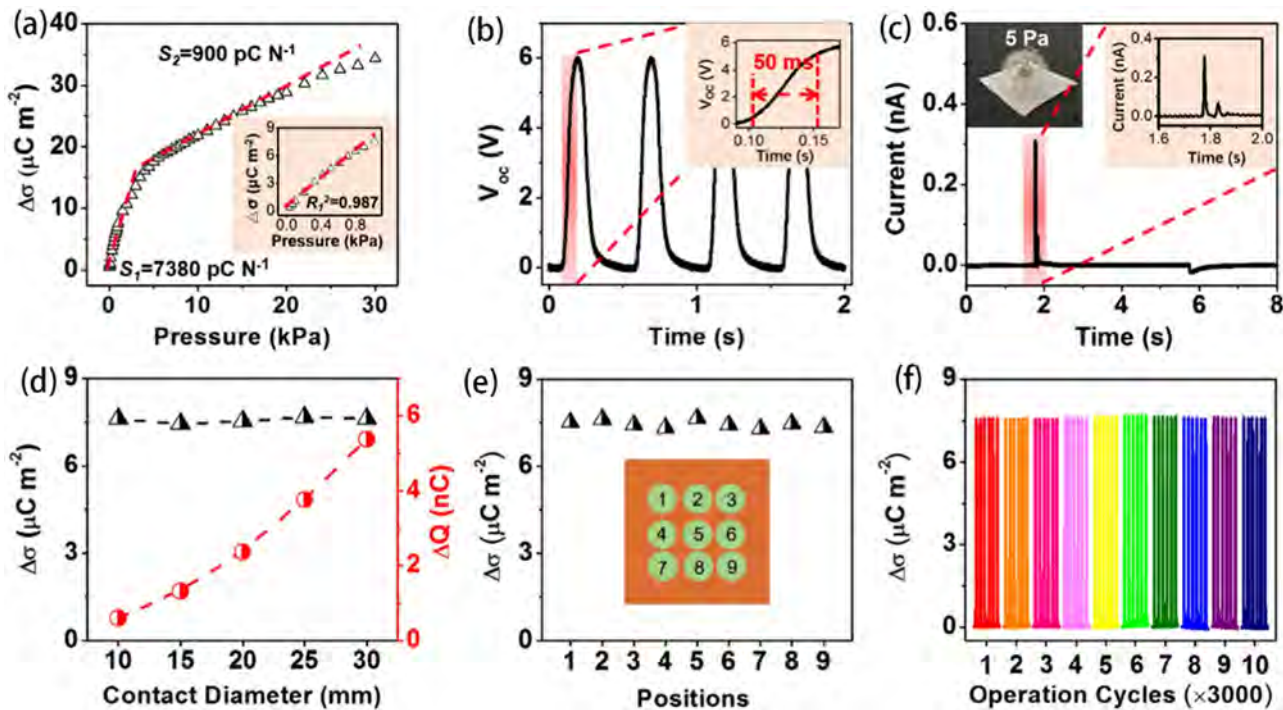


Fig. 18. (a) Transferred surface charge density with increasing pressure. (b) Output voltage of the FPS upon pressure loading/unloading of 1 kPa. (c) Short circuit current response of FPS for a dandelion (~ 5 Pa). (d) Transferred surface charge density (black curve) and transferred surface charge (red curve) of the FPS with different diameters of the contact objects. (e) Distribution of transferred surface charge density when an object (diameter of 10 mm) contacts at different positions on the FPS. (f) The durability of the FPS with a continuous loading/unloading of a periodic pressure of 1 kPa. Reproduced with permission [69]. Copyright 2017, Elsevier.

replacement of power sources also restricts the development of implantable and wearable electronics for both in vitro and in vivo environments. Alternatively, a self-powered pressure sensor has been considered as a promising candidate to solve the power consumption issue for the upcoming era of wearable healthcare sensors.

Piezoelectric pressure sensor is mainly composed of piezoelectric sensitive materials, which can convert mechanical energy and electric energy into each other. Their transduction mechanism could be described as follows: when the material is deformed by external pressure, positive and negative charges separation occurs within the functional material. On the two opposite surfaces of the material, there will appear positive and negative charges arranged in opposite directions, and a potential difference will be formed inside. These potential differences are examined to determine the effect of external forces. The features of piezoelectric sensors in detecting high frequency pressure cause them to be applied to the direction of precision equipment manufacturing.

The flexible piezoelectric pressure sensor has attracted more and more researchers' attention because of its advantages such as convenient material preparation, low cost and convenient electrical signal acquisition. Piezoelectric pressure sensor can directly generate electrical signals in response to mechanical force, which is convenient to realize self-powered sensor system. In addition, the emergence of new piezoelectric materials, including polyvinylidene difluoro-trifluoroethylene (P(VDF-TrFe)), barium titanate (BaTiO_3), lead zirconate titanate (PZT) and zinc oxide (ZnO), has brought a turning point in their development, replacing the traditional brittle ceramics and quartz.

In order to obtain a high-sensitivity piezoelectric pressure sensor, Wang et al. [69] demonstrated a honeycomb-type fluorocarbon piezoelectric pressure sensor (FPS) by using a simple three-step hot-pressing method. By constructing micron-sized voids inside the cell, combined with the excellent charge storage capacity of the fluorocarbon electret, enormous piezoelectric activity can be

achieved. The flexible FPS had significant sensitivity (7380 pC n^{-1}) at low pressure ($< 1 \text{ kPa}$), fast response time (50 ms), very low limit of detection (5 Pa) as well as high stability (30,000 cycles), as shown in Fig. 18.

Lead zirconate titanate (PZT) was a widely used piezoelectric material with high dielectric constant and piezoelectric voltage. However, due to the brittleness of PTZ, processes such as dicing and filling may damage the ceramic, and it is difficult to create complex shapes and device structures. Xie et al. [70] demonstrated a self-powered flexible and highly active pressure and shear sensor based on a freeze-cast ceramic-polymer structure. The layered PZT structure was prepared by freeze-casting, and the piezoelectric composite was formed by impregnation PDMS matrix into the orifice. The structural PZT-PDMS composites had a higher effective longitudinal piezoelectric coefficient (d_{33}^*) of 750 pC n^{-1} , which was higher than that of the whole ceramics due to the combination of bending and bending. Park et al. [72] demonstrated a self-powered flexible piezoelectric pulse sensor based on PZT thin film. The flexible piezoelectric sensor exhibited a sensitivity of 0.018 kPa^{-1} , response time of 60 ms, and good mechanical stability under 5000 pushing cycles.

Lead PZT is a kind of piezoelectric material with good piezoelectric properties, but it will produce heavy metal pollution during sintering, use and disposal, which is harmful to human health. As an environment-friendly material, Barium titanate (BaTiO_3) has been used more and more for vibration energy harvesting due to its excellent dielectric, ferroelectric and piezoelectric properties. However, the inherent mechanical brittleness of inorganic materials makes BaTiO_3 prone to accidental fracture during processing and bonding, which prevents it from forming complex wearable device shapes. Guo et al. [71] designed and developed a wireless flexible wearable piezoelectric device, which was composed of a piezoelectric pressure sensor made of electrospun PVDF/ BaTiO_3 nanowire (NW) nanocomposite fiber. The micro/nano structures acted as active piezoelectric materials, and the use of a polymer

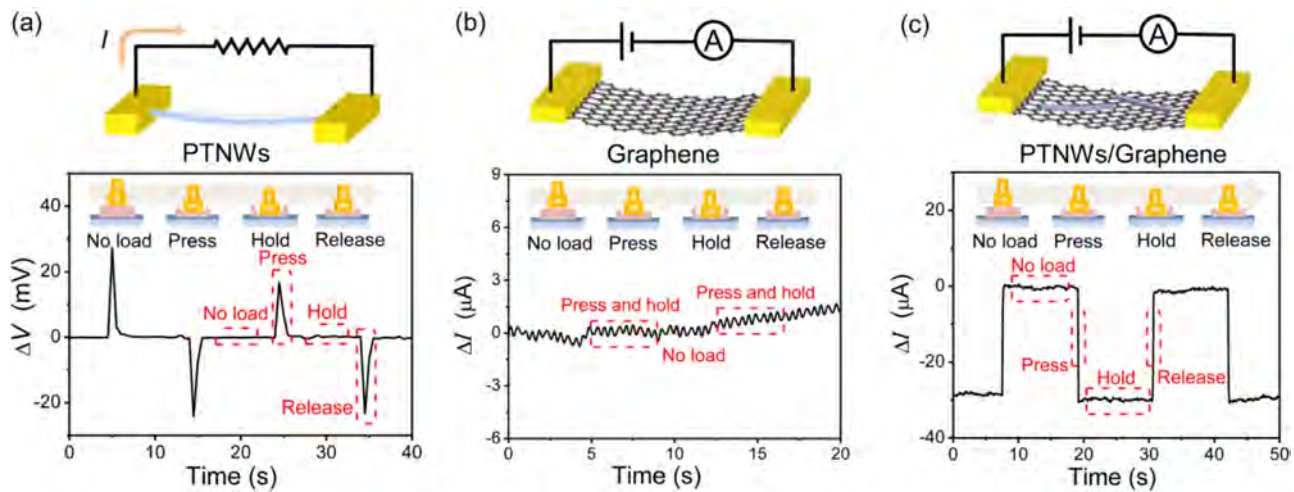


Fig. 19. (a) Pressure response of a pure PbTiO_3 nanowires (PTNWs)-based pressure sensor under a pressure pulse. (b) Pressure response of a graphene-based pressure sensor under a pressure pulse. (c) Pressure response of a PTNWs/G transistor under a pressure pulse. The inset is the dynamic process of the pressure pulse. Reproduced with permission [78]. Copyright 2017, American Chemical Society.

matrix can effectively prevent the entire device from cracking and fracturing under mechanical stress.

Flexible PVDF and P(VDF-TrFE) were an ideal piezoelectric material owing to its favorable chemical inertia, simple manufacturing, and large piezoelectric coefficient. Wang et al. [73] reported on a force sensor based on PVDF fabrics with excellent flexibility and breathability, to be used as a specific human-related sensor. Persano et al. [74] developed a high-performance flexible piezoelectric device based on aligned arrays of P(VDF-TrFE) nanofibers by electrospinning method. The device possessed fast response and high sensitivity, and could detect pressure less than 0.1 Pa, and thus had a great potential in wearable devices. Chen et al. [75] proposed a new nano-template-based electrical growth method to fabricate vertically well-aligned P(VDF-TrFE) nanowire arrays with desired crystals and preferential polarization directions, which were used for vital signs monitoring. In sensing applications, it showed excellent performance. The obtained self-powered flexible sensor had high sensitivity, good stability and strong power generation performance. Under bending conditions, the maximum voltage of the device ~ 4.8 V current density was $\sim 11 \mu\text{A}/\text{cm}^2$. The output voltage of the self-powered sensor was linear to the high voltage and had high sensitivity and piezoelectric voltage. In the P(VDF-TrFE) nanowire array, it was enhanced to 9 times that of the conventional spin coating film. Shin et al. [76] successfully fabricated a highly sensitive, wearable and wireless pressure sensor used zinc oxide (ZnO) nanowires/polyvinylidene fluoride (PVDF) composite membrane. Due to its high dielectric constant, low polarization response time and excellent durability, the hybrid membrane could be used as a real-time pressure sensor to monitor heart rate. The minimum detectable pressure of the mixed membrane was only 4 Pa. Khan et al. [77] proposed a large-area printing flexible pressure sensor developed using full screen printing technology. A 4×4 sensor array was obtained by printing P(VDF-TrFE), its nanocomposites and multi-walled carbon nanotubes (MWCNT) in a parallel plate structure sandwiched between the printed metal electrodes.

Piezoelectric effect has been widely used in dynamic signal detection of pressure sensors. However, these piezoelectric induced pressure sensors present challenges in measuring the static signal generated by the transient flow of electrons in an external load driven by a piezoelectric potential generated by dynamic stress. hen et al. [78] proposed a nanowire/graphene heterostructure pressure sensor for static measurement based on the synergistic mechanism between the strain-induced polarization of the charged piezoelectric nanowires and the variation of carrier

scattering in graphene, as shown in Fig. 19. Compared with traditional piezoelectric nanowires or graphene pressure sensors, the sensor can measure static pressure with sensitivity up to $9.4 \times 10^{-3} \text{ kPa}^{-1}$ and response time as low as 5–7 milliseconds.

Although piezoelectric materials have a wide range of applications in the preparation of flexible sensors, piezoelectric materials also have many obvious characteristics that are not conducive to their application in flexible sensors. For example, piezoelectric materials generally have thermoelectric properties, and electric charges will be generated inside the piezoelectric materials when the temperature changes. In the place where the temperature is prone to change, the strain factor and temperature change of the piezoelectric pressure sensor will lead to the charge change, and the change factor cannot be accurately measured, so the practical application of the piezoelectric flexible sensor still needs further exploration and improvement.

5. Exploration of optimized concept flexible pressure sensor

With the development of pressure sensors, high-performance flexible pressure sensors with high sensitivity, high resolution, rapid response and great flexibility have been extensively studied. But it is far from enough to apply them to practical applications. Herein, we summarize the optimization concepts for the construction of pressure sensing devices toward practical applications.

5.1. Self-powered flexible pressure sensor

In order to reduce power consumption and broaden the use of pressure sensing devices, self-powered pressure sensor have been extensively studied. Piezoelectric effect and triboelectric effect are the two main methods to realize self-powered system. The piezoelectric effect has been introduced in the previous section and will not be repeated here. The self-powered pressure sensor based on triboelectric effect has been extensively researched due to its simple structure, low cost, easy manufacturing and integration with other processing technologies.

In 2012, a new ambient energy harvesting technology named triboelectric nanogenerator (TENG) was developed based on the coupling of electrification effect and electrostatic induction [79]. TENG can be categorized into four main types according to different operating principles, including contact-separation (CS) mode,

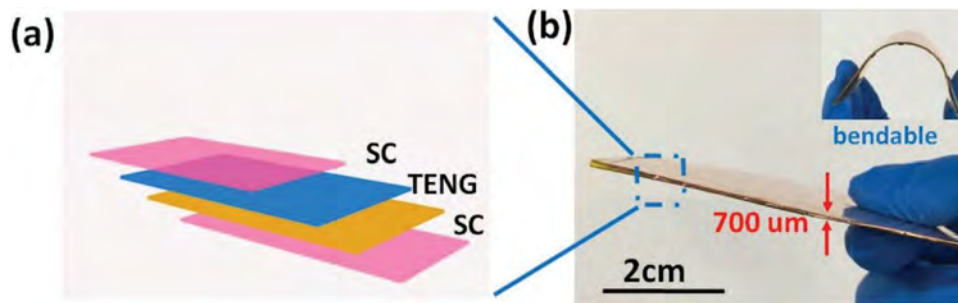


Fig. 20. Schematic illustration of biomimetic pressure sensor. a) Architecture of the pressure sensor. b) Photograph of pressure sensor. Reproduced with permission [81]. Copyright 2017, Wiley-VCH.

relative-sliding (RS) mode, single-electrode (SE) mode and free-standing (FS) mode [80]. Among them, contact-separation mode and single-electrode mode are widely used for pressure sensing. Fan et al. [80] developed the first flexible nanogenerator for pressure sensing based on a contact-separation model, which was optimized by using a transparent polymer material and a micropatterned surface to sense the pressure of water droplets and falling feathers. Zou et al. [81] designed a biomimetic pressure sensor based on ultrathin super-capacitor (SC) and flexible TENG, as shown in Fig. 20. With a sandwich design, the triboelectrodes sandwiched within the two solid super-capacitors could not only store electrical energy by a wireless energy transfer mode but also help TENG imitate the receptor's unique characteristics by simultaneously measuring both static and dynamic pressures in a self-driven mode. A self-powered flexible tactile sensors fabricated by Zhu et al. [82] was based on contact electrification. Enabled by the unique sensing mechanism and surface modification by polymer-nanowires, the triboelectric sensor showed an exceptional pressure sensitivity of 44 mV/Pa ($0.09\% \text{ Pa}^{-1}$) and a maximum touch sensitivity of 1.1 V/Pa ($2.3\% \text{ Pa}^{-1}$) in the extremely low-pressure region ($<0.15 \text{ kPa}$). Chen et al. [83] used flexible hollow microstructures to enhance nanogenerators. They first came up with a contactless heartbeat and breathing monitoring system that can work at high pressure weight without direct contact with the skin. Using only commercial polymer films, a high dynamic pressure sensitivity of $18.98 \text{ V} \cdot \text{kPa}^{-1}$ and a wide operating range of 40 kPa can be achieved simultaneously. The special advantages of physiological testing under high pressure were also observed. In the non-contact mode, continuous and reliable heartbeat and respiratory information was successfully detected and transmitted to the mobile phone.

Relying on the unique pressure response characteristics of the open-circuit voltage and short-circuit current, Lin et al. [84] implemented static and dynamic pressure sensing on a single device, as shown in Fig. 21. The active triboelectric sensor has a high sensitivity of 0.31 kPa^{-1} , ultra-fast response time of $<5 \text{ ms}$, long-term stability (30000 cycles) and a low detection limit of 2.1 Pa. Meng et al. [85] reported a flexible weaving constructed self-powered pressure sensor (WCSPS) for measurement of the pulse wave and blood pressure in a noninvasive manner. The WCSPS hold an ultra-sensitivity of 45.7 mV Pa^{-1} with an ultrafast response time of less than 5 ms, and no performance degradation was observed after up to 40 000 motion cycles. Das et al. [86] developed a self-powered triboelectric pressure sensor using laser-ablated graphene for self-powered human gesture detection (SP-HGD) and wearable healthcare applications. The sensor with micro-structured electrode had higher sensitivity (7.697 kPa^{-1}), lower detection limit ($\sim 1 \text{ Pa}$), faster response time ($<9.9 \text{ ms}$), and higher stability over 4000 compression release cycles. The method was suitable for adaptive preparation steps in self-powered systems, especially in electronic skin and healthcare applications, at a very low cost.

5.2. Multifunctional flexible pressure sensors

For applications in e-skin, robotics, and human-machine interfaces, pressure sensing, as well as temperature, humidity, and other stimulus sensing are necessary. Decoupling signal analysis and integrating multi-functional features into a sensor array is the main challenge of multi-functional pressure sensors. In this part, we have summarized some flexible pressure sensors with multifunction.

Optical and electrical dual-mode sensors are obtained by integrating pressure sensors and mechanical luminescence sensors. Wang and Pan et al. [87] demonstrated a self-powered optical and electrical dual-mode sensing by integrating triboelectric and mechanoluminescent sensor matrix, which could realize wide range dynamic pressure sensing but had limitation in static pressures detection. Zhang et al. [53] introduced a dual-mode pressure sensor as an electronic skin to distinguish and map different levels of dynamic and static pressure, as shown in Fig. 22. The electronic skin integrated two sensing components with complementary pressure ranges. Under low pressure ($<5 \text{ kPa}$), the electronic skin exhibited a highly sensitive capacitive response via the microstructural PDMS. When in a relatively high pressure state ($>60 \text{ kPa}$), the phosphor particles embedded in the PDMS matrix could be triggered by the enhanced electric field. The bright luminescence made the pressure profile immediately visible, and the luminous intensity was readable like the naked eye. The two different induction modes were electrically and optically simulated to mechanoreceptors and nociceptors in biological skin, respectively. Their sensitivity to soft touch and pain pressure reached 0.66 kPa^{-1} and 0.044 kPa^{-1} , respectively. In addition, the stretchable elastomer matrix and transparent electrodes enable the electronic skin to withstand large mechanical deformation, indicating its potential applications in robotic body coverage.

Temperature sensing is also the basic function of next-generation artificial intelligence products, whereas it is still a challenge to use a single device to sensitively detect temperature and pressure at the same time. Zhang et al. [88] demonstrated temperature–pressure dual-parameter sensors utilizing microstructure-frame-supported organic thermoelectric (MFSOTE) materials. Used the independent thermal voltage resistance effect in a single MFSOTE device, the external stimulus was transformed into an independent electrical signal to realize the simultaneous monitoring of temperature and pressure. Effectively converted temperature and pressure stimulation into two independent electrical signals, allowed instant sensing of temperature and pressure. Accurate temperature resolution was less than 0.1 K, and high pressure sensing sensitivity was as high as 28.9 kPa^{-1} , as shown in Fig. 23. What's more, these dual parameter sensors could be self-powered and have good sensing performance. Yoon et al. [89] reported a simple fluid capacitive sensor based on microfluidic technology, which could achieve multi-mode sensing performance. Used ionic liquid as microchannel electrode and CNT/PDMS com-

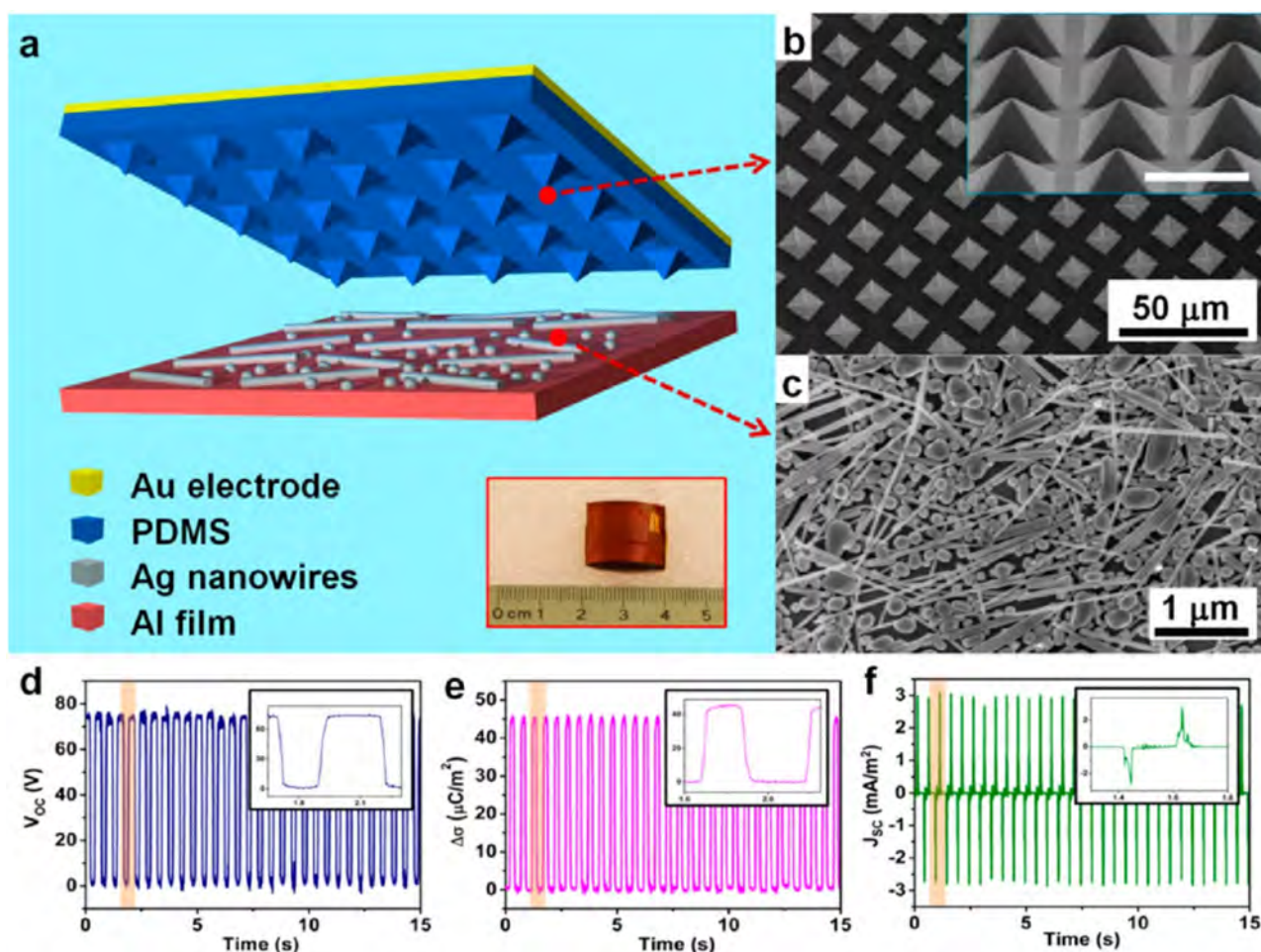


Fig. 21. The basic structure and typical electrical response of the triboelectric active sensor applied with a cyclic pressure. (a) The schematic illustration of the TEAS device with both inner surfaces modified by micropattern PDMS structures and Agnanowires/nanoparticles composite. The inset is a photograph of the TEAS device. (b) The SEM image of the pyramid-structured micropatterns fabricated on the inner surface of the PDMS membrane. The inset is a high magnification SEM image of the micropatterns, and the inset scale bar is 10 μm . (c) The SEM image of the Ag nanowires/nanoparticles composite assembled on the inner surface of the Al foil. (d,e) The measured typical electrical response including (d) the open-circuit voltage, (e) the transferred charge density, and (f) the short-circuit current density of the TEAS upon a cyclic pressure. Reproduced with permission [84]. Copyright 2013, American Chemical Society.

posite as dielectric layer, a multifunctional microfluidic capacitance sensor was realized. Microfluidic capacitive sensors had excellent sensor performance and could detect local pressure and even side pressure changes and ambient temperature changes. What's more, a gas-permeable and stretchable electrode with the conductive nanomesh structure was fabricated through electrospinning and depositing, which could be directly adhered to human skin without inflammation. By integrating flexible and thin polymeric temperature sensor with positive temperature coefficient as well as pressure-sensitive rubber, the on-skin sensors were able to detect pressure and temperature simultaneously [90].

5.3. Self-healing flexible pressure sensor

In recent years, self-healing hydrogels with good mechanical properties and adhesion have attracted great interest because the self-healing ability can extend the service life of materials, and has shown application value in self-healing electronic equipment and biomedical fields.

The first self-healing pressure sensor was developed by Bao et al. [91], which was manufactured using supramolecular organic polymers to form a hydrogen bond network embedded in nickel nanostructured particles. On rupture, the initial conductivity was repeatable restored with $\sim 90\%$ efficiency after 15 s healing time,

and the mechanical properties was completely restored after ~ 10 min. The external pressure may change the space of the nickel particles and their resistance at the same time, thus obtaining the piezoresistive properties of the composites. The pressure sensor is obtained by sandwiching the piezoresistive composite material between the layers of the conductive composite material using a parallel plate structure, as shown in Fig. 24. In order to improve transparency, Jing et al. [92] synthesized a polyvinyl alcohol/cellulose nanofibril (PVA/CNF) hydrogel with dual-crosslinked networks for highly transparent, stretchable, and self-healing pressure and strain sensors. The developed hydrogel had a moderate modulus of 11.2 kPa, and a high elongation rate of 1900 %. It spontaneously self-heals within 15 s upon contact without any external stimuli, had a high transmittance of over 90 %. Xu et al. [93] fabricated the sodium casein-polydopamine (SC-PDA) hydrogel. The SC-PDA hydrogel showed stretchability and self-healing ability. The fracture stress of the hydrogel was 170 kPa, the fracture strain exceeds 2100 %, and it had excellent reversible adhesion properties on various materials and even human skin. In addition, because of the existence of sodium ions, the SC-PDA hydrogel had a sensitive deformation-dependent conductivity, which can act as a flexible strain and pressure sensor. Depended on the self-healing polysiloxane undergoes a solid-liquid-solid transition during the self-healing process. Zhao et al. [94] reported a self-healing pres-

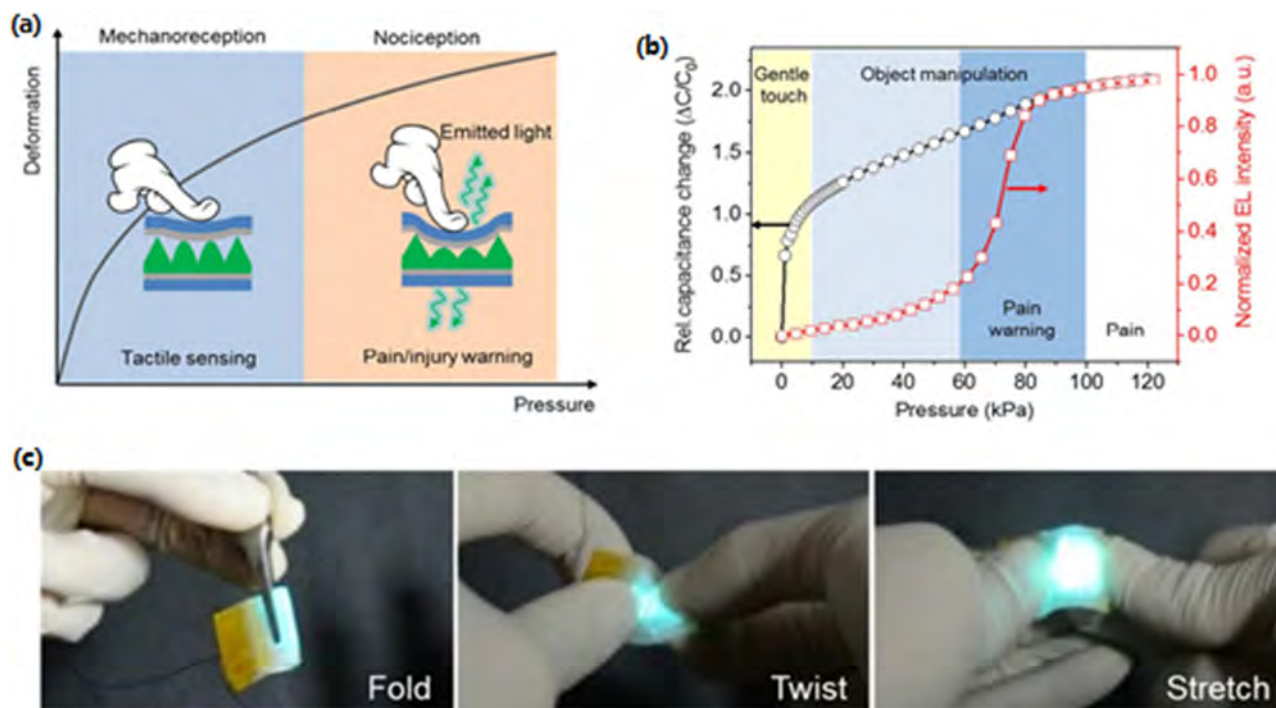


Fig. 22. (a) Illustration of the nonlinear deformation of the e-skin with the increase of pressure and insets schematically show different sensing modes in different pressure ranges. (b) Capacitive and luminescent responses of the e-skin to different pressures. (c) Luminescent pictures of folded, twisted and stretched e-skin. Reproduced with permission [53]. Copyright 2017, American Chemical Society.

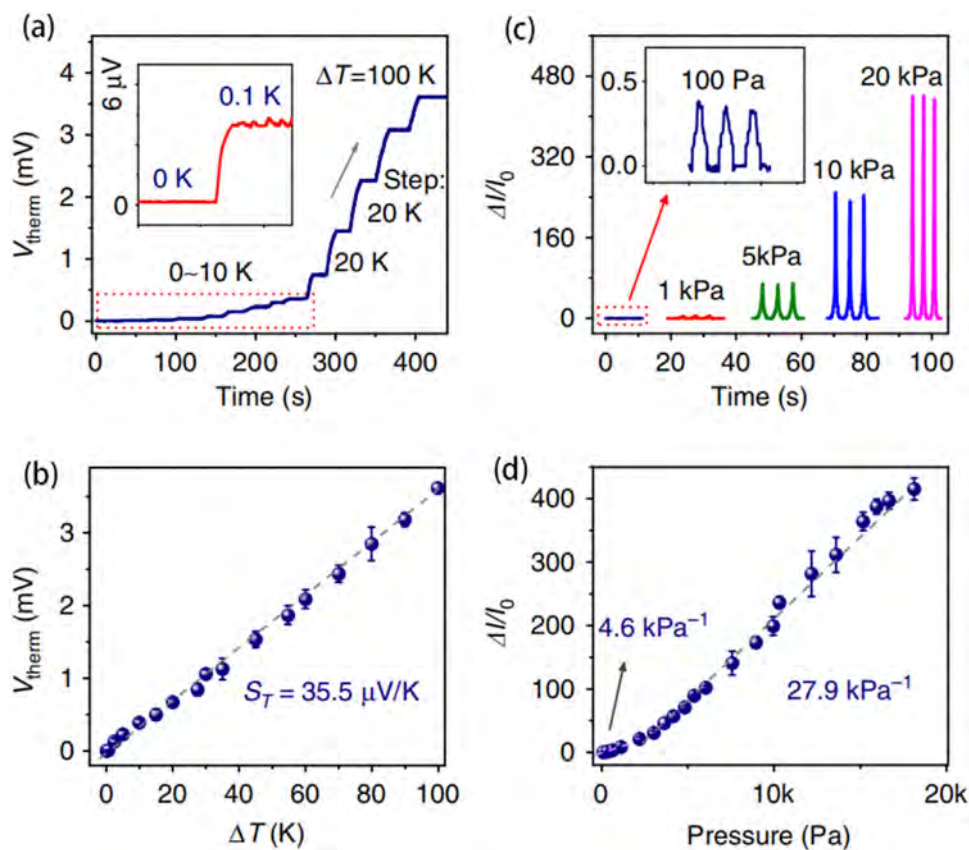


Fig. 23. (a) Output voltage of a MFSOTE device to a biased temperature gradient range of 0–100 K. The insert shows the magnified response signal of a MFSOTE device to a temperature gradient of 0.1 K. (b) Measured output voltage as a function of temperature gradient. (c) Current response of a MFSOTE device to various pressures at a constant voltage of 0.1 V. The insert shows the magnified response signal to the pressure of 100 Pa. (d) Current responses of the MFSOTE devices to various pressures. Reproduced with permission [88]. Copyright 2015, Springer Nature.

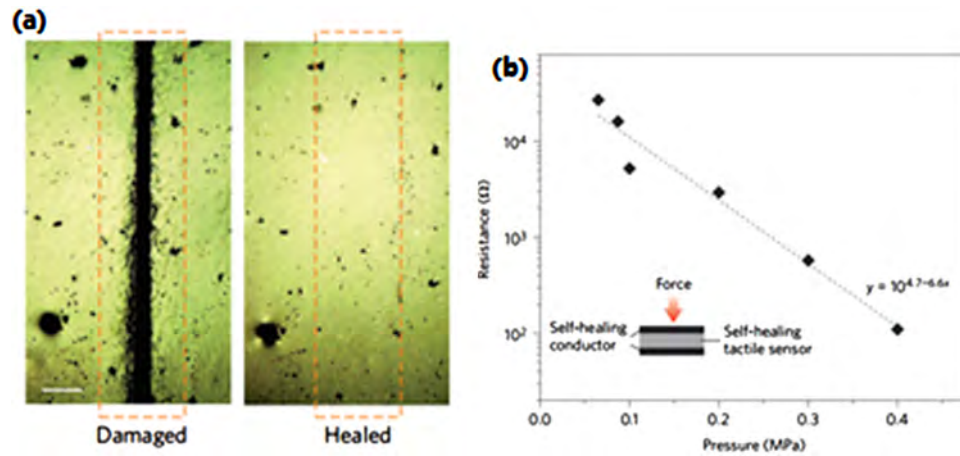


Fig. 24. (a) Optical microscopy images of a damaged sample and complete scar healing. (b) LED eyes light up after the elbow is bent. Reproduced with permission [91]. Copyright 2012, Springer Nature.

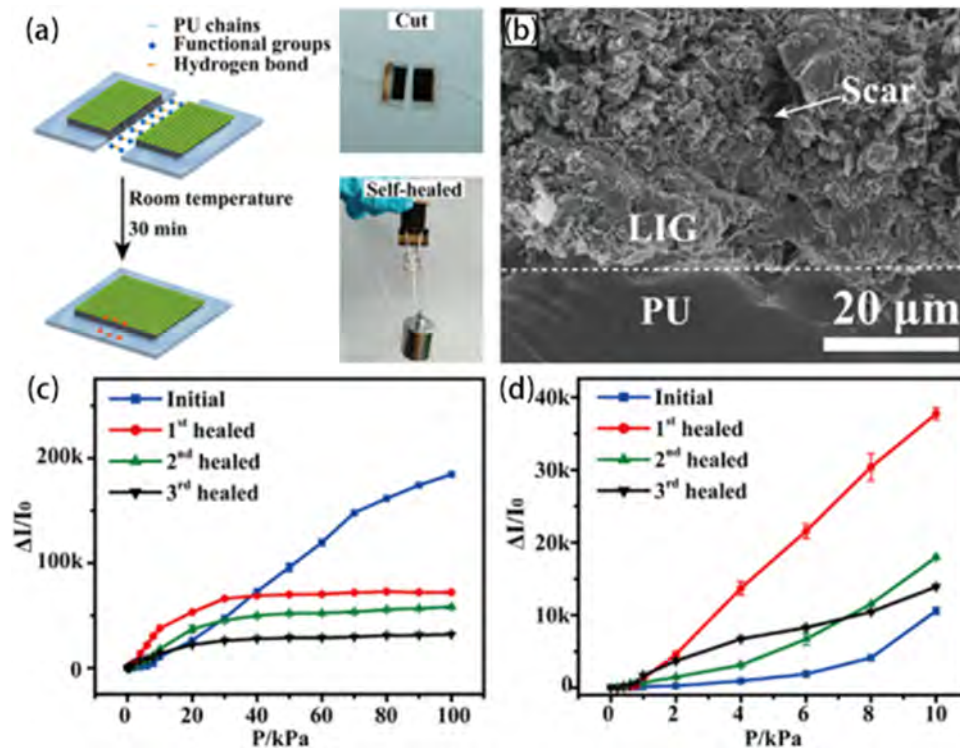


Fig. 25. Self-healing performance of the sensor with 1.0 mg to 1.3 μm PS layer. (a) Illustration of the self-healing process. (b) SEM image showing the cross section of a self-healed LIG and PU layer. (c) Relative current change performance of sensor after self-healing, each for 30 min at room temperature. (d) Magnified view for the pressure range of 0–10 kPa in the plot (c). Reproduced with permission [95]. Copyright 2020, American Chemical Society.

sure sensor based on a polysiloxane network cross-linked through dynamic Diels-Alder bonds. The tensile stress of nanocomposite containing 35 wt% graphene was 1.09 MPa, which was more than 1700 % higher than that of the elastomer, indicating that the tensile properties and tensile stress were significantly improved. The self-healing pressure sensor had a high sensitivity of 0.765 kPa^{-1} . Recently, inspired by the bean pod structure, Tian et al. [95] presented a flexible pressure sensor architecture consisting a microspacer core layer of polystyrene (PS) microspheres, sandwiched between two laser-induced graphene/polyurethane (LIG/PU) films. The improved linearity of the pressure sensor provides a wide sensing range of up to 100 kPa with excellent stability (over 1,000 load-unload cycles) and sensitivity of 149, 659 and 2048 kPa^{-1} , respectively, in the pressure ranges of 0–1, 1–10 and

10–100 kPa. Furthermore, after three cut-and-heal cycles at room temperature, the severely damaged device was able to self-heal and maintain high sensitivity, as shown in Fig. 25.

6. Conclusions and outlook

In the past few years, researchers have made great progress in the sensitivity, response time, detection limit and other performance of flexible pressure sensors by developing various novel materials and optimized structures. In this review, we introduce the recently major progress in flexible pressure sensors from the basic design of three sensing mechanisms to optimization concept for practical application. The newly emerging materials and structures used in fabrication of the piezoresistive, capacitive, and piezoelec-

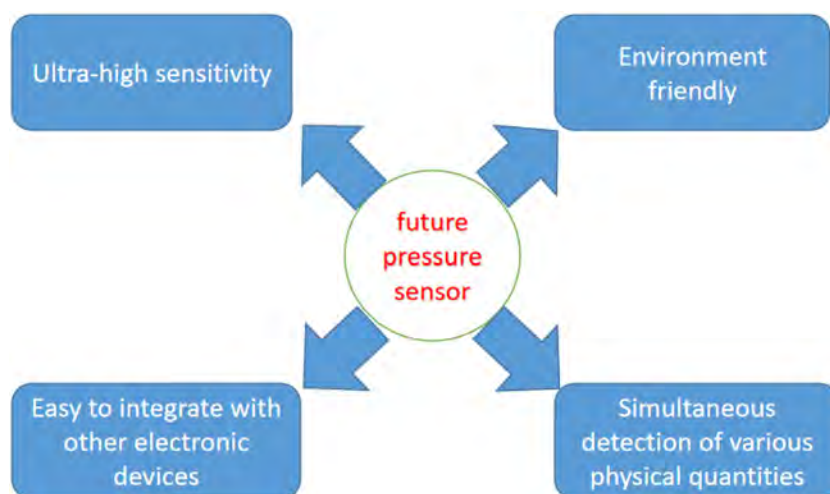


Fig. 26. The development trend of flexible pressure sensors in the future.

tric pressure sensors are comprehensively illustrated. In addition, high-performance flexible pressure sensors designed for different application requirements such as self-powered pressure sensors, multifunctional pressure sensors, and self-healing pressure sensors are also discussed. The tremendous progress made by these flexible pressure sensors and their potential applications will become an indispensable part of the next generation of electronic products.

As shown in Fig. 26, according to the application demands of wearable devices, artificial intelligence, biomedicine and other fields, the development trend of sensors in the future is mainly in the following aspects.

- (a) Ultra-sensitivity is a necessary trend for the future development of pressure sensors. Although many pressure sensors with high sensitivity have been developed, the sensitivity of the pressure sensors developed is not enough for some special-demand applications.
- (b) The preparation process of pressure sensors will inevitably produce some substances harmful to the environment. It is urgent to find a cheap and convenient preparation method and high-performance and environmentally friendly functional materials.
- (c) Pressure sensors need to be integrated with other flexible devices and can be combined with big data of the Internet of Things to conform to the upcoming development trend. Therefore, another big challenge is to integrate the flexible wearable pressure sensor with the signal transmission, data processing unit, power supply and performance optimization strategy that is highly maneuverable.
- (d) In order to meet the application requirements of electronic skin, wearable electronic devices and other fields, it is very important to develop pressure sensor arrays that can detect various physical quantities simultaneously. At present, most of the pressure sensor arrays with two or more physical quantities detecting will not exclude the interaction between physical quantities. For instance, temperature has great influence on any other physical quantities, however measuring temperature is an indispensable ability of electronic skin. New active materials, structure designs and transduction principles are looked forward to achieve this application requirement.

Therefore, although there has been a lot of research studies on wearable pressure sensors, there are still a lot of challenges and breakthroughs to explore further.

Declaration of Competing Interest

The authors report no declarations of interest.

Acknowledgements

This work was supported by Jiangsu Natural Science Foundation for Excellent Young Scholar (BK20170101), and the Scientific Research Foundation of Nanjing University of Posts and Telecommunications (NY220011, NY220034). Natural Science Foundation of Jiangsu Higher Education Institutions of China (Grant No. 20KJA510001), and the Postgraduate Research & Practice Innovation Program of Jiangsu Province (SJCX19.0270).

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