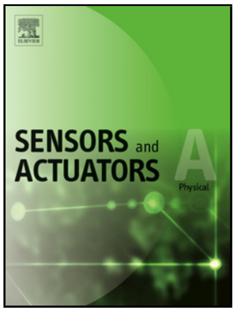

Research progress of flexible capacitive pressure sensor for sensitivity enhancement approaches

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Research progress of flexible capacitive pressure sensor for sensitivity enhancement approaches

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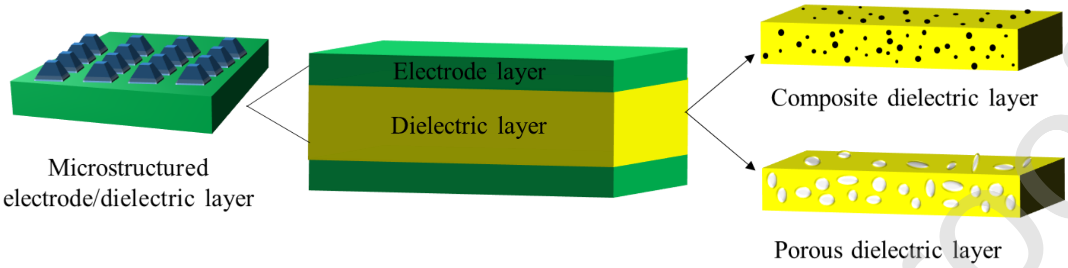
Graphical abstract



In this paper, three methods to improve the sensitivity of flexible capacitive pressure sensors are mainly reviewed, including (1) constructing microstructure of the dielectrics or electrodes, (2) adding conductive fillers to polymer elastomer to generate a composite dielectric, (3) introducing micro-holes into the dielectric layer.

Highlights

1. The main factors affecting the capacitance are dielectric constant and distance of electrode during compression.
2. Microstructure and porous materials mainly enhance the capacitance change by reducing the Young's modulus.
3. The composite dielectric improve the sensitivity by increasing the relative dielectric constant.

Abstract

Flexible pressure sensors have played a great role in acquiring information from human and automatics because of their wide use in electronic skin, soft robot, human-machine interaction and so on. Among a variety of flexible pressure sensors, capacitive pressure sensor has many advantages like simple structure, insensitive to temperature and humidity, low power consumption, etc. It is easy to fabricate such kind of pressure sensor, nevertheless, how to improve its sensitivity to broaden the high effective application has been a hotspot issue in recent years. In this paper, a large amount of research outputs on sensitivity improvement have been reviewed for flexible capacitive pressure sensor, including the aspects from introduction of performance evaluation indicators, working principle, generally used materials and capacitor structures to the methods of how to improve the sensitivity of capacitive pressure sensors. Then, the effective ways to obtain high sensitivity of pressure sensors have been compared and the development trend of flexible capacitive pressure sensor is prospected. This paper aims to provide references for the further research on the efficient fabrication of flexible capacitive pressure sensors and effective usage of such sensors in high sensitivity requirements of application areas.

Keywords: Capacitive pressure sensor; Dielectric; Electrode; Flexibility; Sensitivity;

1. Introduction

Pressure is a physical vector that forces an object to contact the surface of another one tightly, leading to the surface spatial deformation or performance variation more or less. Pressure sensor is a kind of electronic device that can convert the pressure signals into the corresponding electronic signals [1]. To date, a few traditional mechanical sensors have been reported with an accurate measure of a variety of pressures along with the development of technology. However, they still indicate some demerits like large volume and heavy weight that are difficult to meet the requirements of modern micro pressure sensors, especially for wearable electronics such as their portability, flexibility and adaptability [2, 3].

Flexible pressure sensors have many advantages over stiff sensors. Some novel flexible sensors were reported with light weight, high flexibility, good resolution and fast response [3, 4], which show great potential use in electronic skin (e-skin) [5-8], wearable devices [9], etc. Here, a case of a flexible pressure sensor with high sensitivity is particularly attractive and important to some researchers as it can measure rather weak pressure signals such as human blood pulse, heartbeat and breathing. Furthermore, pressures inside human body such as brain, blood, kidney pressures, etc. may give people very important information on health for in time warning of major diseases [10].

Regarding flexible sensors, in fact, the components and structure parameters would determine the sensing performances. Moreover, some indicators like sensitivity are also important to judge the quality of flexible pressure sensors [11, 4]. According to the sensing mechanism, flexible pressure sensors mainly have three categories, i.e., capacitance [12,13], piezoresistivity [14] and piezoelectricity [15]. In comparison with piezoresistive and piezoelectric types, the capacitive pressure sensor shows superiorities of good stability, simple structure and low power consumption [16]. However, the capacitive pressure sensors are difficult to achieve high sensitivity due to the limitation of Young's modulus of elastic dielectric layer [4]. The measurement accuracy of such sensor is increasingly required to expand the application range of the sensor. Therefore, many researchers have launched a series of studies on the improvement of sensitivity of capacitive flexible pressure sensor.

This paper aims to review the most advances of flexible capacitive pressure sensors for mainly the study of improvement methods of sensitivity. This includes the aspects of capacitor structure, flexible materials, working principle and performance indicators for the sensitivity enhancement methods. In this paper, the preparation materials and working principle of capacitive pressure sensors are firstly introduced. Then, the most advances on improving the sensitivity of capacitive flexible pressure sensors in recent years are analyzed. The development trend and application prospect of flexible pressure sensors are summarized and discussed finally.

2. Flexible pressure sensors

2.1 Classification

On the basis of sensing range, there are four kinds of flexible pressure sensors that can measure the pressure ranges of ultra-low-pressure (<1 Pa), subtle-pressure (1 Pa - 1 kPa), low-pressure (1 kPa - 10 kPa), medium-pressure (10 kPa -100 kPa) and high-pressure (>100 kPa) respectively, as shown in Fig. 1. The commonly encountered human motions such as gentle touching and object handling are mainly in the low (1 kPa - 10 kPa) and medium (10 kPa-100 kPa) pressure ranges [17]. In contrary, the pressure from sound and breathing of human body belongs to the ultra-low pressure (less than 1 Pa), and is difficult to detect using traditional pressure sensors. Therefore, the ultra-sensitive pressure sensor with low detection limit have far-reaching significance for developing products like hearing aids, microphones and so on [10]. Furthermore, the sensor has a great potential use in the highly sensitive e-skin owing to an ability to detect slight pressure changes (from 1 Pa to 1 kPa), exhibiting even a better perception than human skin [10]. Besides, the sensor can also be used for wearable touch keyboards [18], highly sensitive touch screens, etc.

The low pressure (range from 1 kPa to 10 kPa) is commonly encountered in human life, which is equivalent to the gentle touch between peoples [17]. For this pressure range, flexible sensors are often used for detecting some scenarios like blood pressure, intraocular pressure, pulse and bladder pressure. Therefore, such sensors can be widely used in medical monitoring. For medium pressure range (10 kPa-100 kPa), in similarity to the pressure of objects operated by hands, some sensors can be used to detect the pressure or movement [19]. Pressure sensors for the high-pressure range (>100 kPa) are usually used in some special scenarios, such as industrial robots, colonoscopes, prostheses [20]. For such scenarios, the sensor’s role is mainly to sense the critical pressure to avoid damage caused by collision or contact, especially in the application of robot, which can avoid collision with objects and perceive the surrounding environment better.

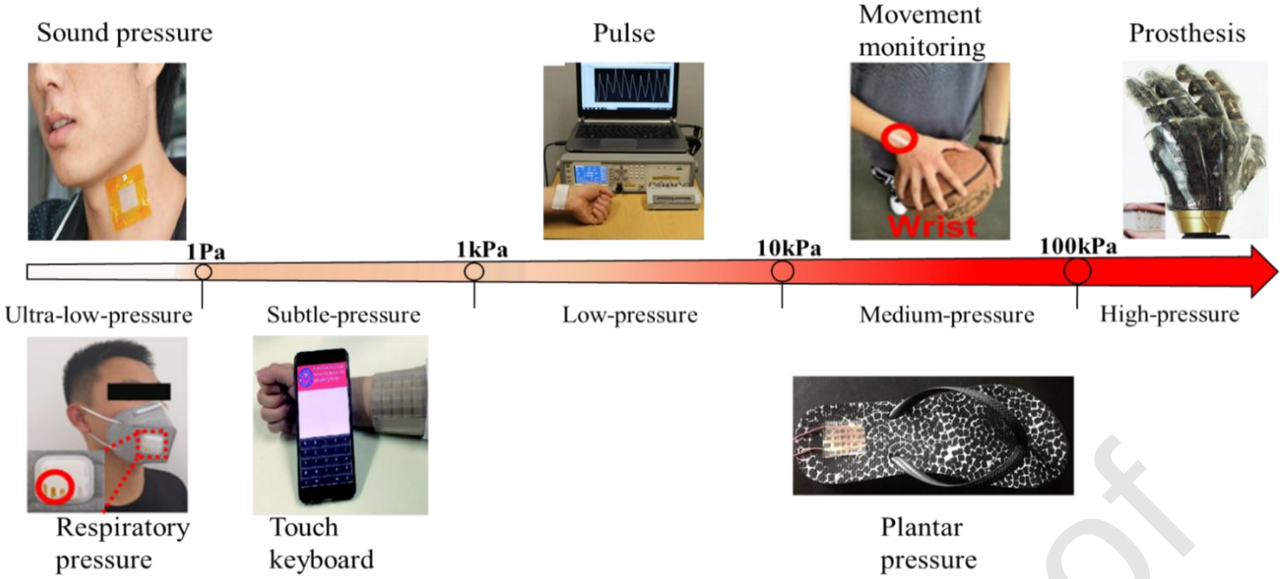


Fig. 1. Pressure classification and application of pressure sensors [10, 18, 21, 22, 23, 24, 25].

2.2 Evaluation indicators

A flexible pressure sensor is a kind of flexible device that is able to recognize the physical spatial deformation such as bending, winding or twisting. The flexibility of the sensor can be constructed in terms of the perspectives of materials, structures and applications. Here, a flexible pressure sensor is normally evaluated by a few key parameters including linearity, stability, limit of detection, response time and sensitivity.

In practical, the signal output of a sensor is a characteristic curve. Linearity is a parameter usually expressed as the maximum deviation in percentage between the characteristic curve and the fitted straight line. The smaller the value means the higher the linearity of the sensor's characteristic curve. The linearity has a certain influence on the sensing range of the sensor. A wider linear sensing range gives more accurate and reliable value obtained by the sensor. Therefore, it is necessary to choose the sensor with a large linear sensing range as far as possible [10]. Stability refers to the ability of a sensor to maintain its original performance after use for a period of time, which is also one of the important factors to determine the life cycle of sensor. Limit of detection is defined as the minimum detectable pressure from the output signal of sensor which is also an extremely important parameter in the acquisition of ultra-low-pressure such as sound waves and pulse. The smaller value of the limit of detection indicates the higher ability of the sensor to sense small pressures. Response time is the time interval from an applied pressure on a sensor to give the output signal, which is very useful in dynamic pressure monitoring. The response time reflects the response speed of a sensor to any applied pressure, and determines the real-time performance of a sensor in acquiring pressure signals. With the advances of technology, some flexible sensors have achieved the instant response time in less than 100 ms [10]. Sensitivity is one of the most important performance indicators for all pressure sensors which will be reviewed and discussed in detail in the following section.

3. Flexible capacitive pressure sensors

Flexible capacitive pressure sensor is an important type of flexible sensor in detecting loading. This section will review the research progress on such pressure sensor, including how to fabricate it, the working principle and the performance evaluation indicators.

3.1 Sensor materials and structures

A capacitive sensor derives from the deformation of a capacitor [3], which is composed of a pair of conductive electrodes and a middle layer of dielectric substance. A typical flexible capacitive sensor is a sandwich-like structure that is composed of a pair of flexible electrodes and a flexible dielectric middle layer [26]. In order to ensure the good flexibility, the electrodes are usually made of flexible substrates and conductive materials according to the required properties of the sensors.

(1) Flexible substrates

In comparison with the rigid metal and semiconductor materials used in traditional pressure sensors, flexible substrates in pressure sensors are suitable for more applications. After review of research works, it is found that the polymer films such as polydimethylsiloxane (PDMS), polyester (PET), ecoflex, polyurethane (PU), polyimide (PI) and polyvinyl alcohol (PVA), etc., are commonly used as flexible substrates. Among them, PDMS is a widely used substrate in sensors for its high elasticity, thermal stability and chemical stability. Besides, fabrics with more intergaps and good flexibility are also applied as flexible substrates. Min et al. [27] plated Ni-Cu-Ni metals on a polyester fabric as electrode layer and assembled it with 100% polyester woven fusible interlining to generate a flexible capacitive sensor. The sensor was demonstrated with excellent sensing performance and was successfully utilized to detect human respiration. Fig. 2(a) shows the variations of the distance and relative area between two electrodes caused by the force from the movement of abdominal and chest muscle in inhalation, leading to the change of capacitance. In order to verify the practical application of the sensor, an experimental device was built for the respiration detection, as shown in Fig. 2(b). The respiration signal was detected by a belt type textile capacitive respiration sensor (BTCRS) and a nasal thermocouple at the same time. Fig. 2(c) and (d) show the results of respiration detection by the BTCRS and nasal thermocouple. Fig. 2(c) compares the respiration rate from the two methods, reflecting a similarity between the two different methods graphically. It can be seen from the figure that the respiratory rate measured by BTCRS is highly correlated with that measured by the other method ($R=0.9846$, $p<0.001$). Fig. 2(d) is the Bland-Altman analysis that compares the measured results with the reference value. In the work, the bias is -0.0015 bpm, the standard deviation is 0.2568 bpm, and the limit of the agreement is -0.5049 to 0.5018 bpm. It is noted from the comparison that the detection result of BTCRS is very similar to the reference value that was measured by nasal thermocouple, indicating that the prepared sensor can be applied to some practical applications.

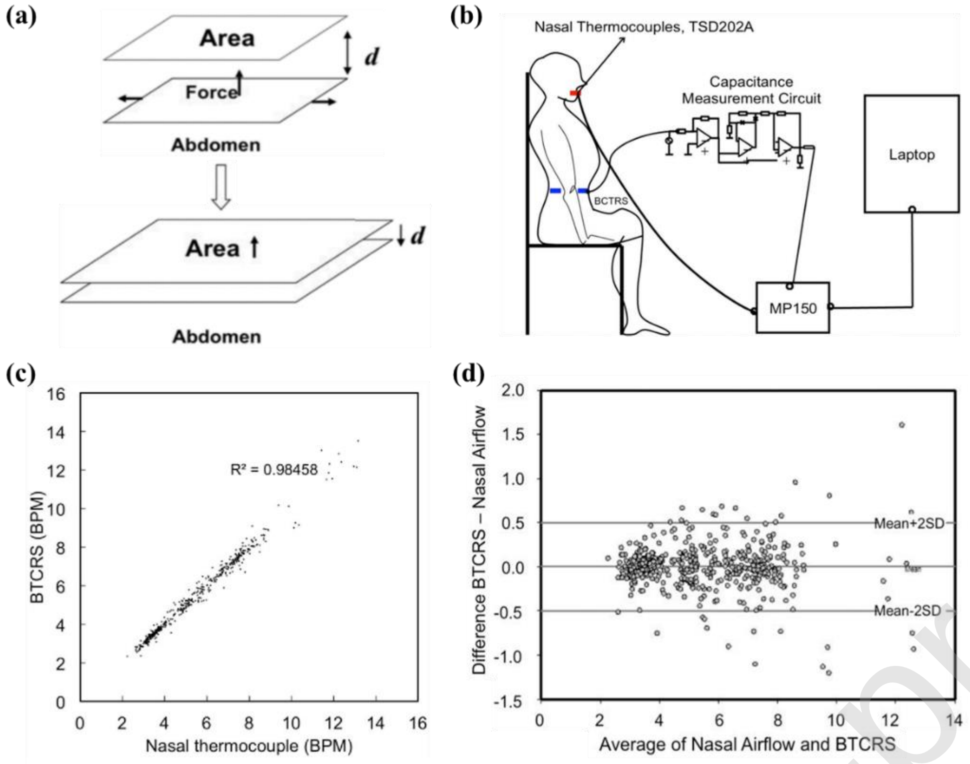


Fig. 2. Respiratory detection characteristics of the sensor and schematic diagram of, (a) the capacitance change during inhalation, (b) experimental device for respiration detection; (c, d) comparison of respiration detection results measured by a belt type textile capacitive respiration sensor (BTCRS) and a nasal thermocouple.

(2) Electrode materials

Conductive electrode is the core of a flexible sensor. In addition to the good conductivity, other factors such as chemical stability and mechanical properties should be considered for selection of electrode materials. Currently, metal films [28, 29], metal nanowires [30, 31] and carbon materials (carbon nanotubes [32, 33], graphene [34], carbon-black [35]) are commonly used as conductive materials in electrodes. Besides, polymer gel films are also utilized as sensor conductive materials. For example, Sun et al. [36] prepared a double-layer capacitor by using the ionic gel film as the conductive material, which has manifested a greatly improved sensitivity of the sensor.

(3) Dielectric materials

In order to improve the sensitivity of sensor, a material with a high dielectric constant is usually applied in dielectric [37]. For a flexible capacitive pressure sensor, the dielectric layer materials can be roughly divided into two types, polymer dielectric and fabric dielectric. The former is similar to a polymer film, such as PDMS, PU, organosilicon elastomer, polystyrene, PET, etc. In order to improve the sensing performance, sometimes, it is necessary to change the structure of the polymer dielectric materials. After all, the Young's modulus of the commonly used polymer dielectric is relatively high [38]. Then, a porous dielectric layer may be favored in fabrication of a pressure sensor, for instance, a fabric dielectric. After a review, it is found that the fabric dielectric mainly has warp-knitted spacer type and woven structure type [39]. For the two fabrics, the spacer fabric is generally thicker with a high range of deformation. Therefore, the detection range is relatively higher for the sensor with a spacer structure dielectric layer.

3.2 Working principle of capacitive pressure sensor

The working principle of a capacitive pressure sensor is to sense the change of capacitance during an external loading on it. Here, a typical capacitive pressure sensor may be the type of variable distance or the type of variable area between the two layers of electrodes, as shown in Fig. 3(a). When an external load is applied to a capacitive sensor, the change of dielectric layer can be recognized, i.e. the change of distance or area of electrodes, thus reflect the loading value. It can be seen that the dielectric has a great influence on the sensing ability. As is known, the capacitance equation is as follows,

$$C = \frac{\varepsilon_0 \varepsilon_r A}{d} \qquad (1)$$

Where *C* is the capacitance of the sensor, ε_0 is the vacuum dielectric constant, ε_r is the relative dielectric constant, *A* and *d* represent the relative area and distance between two electrodes respectively. As shown in Eq. (1), there are many factors that influence the *C* value individually or simultaneously. However, the results of many studies indicate that the main factors affecting the *C* value are parameters of ε and *d* during pressure induced compression, which endows the optimal design of pressure sensor with a more clear direction [7].

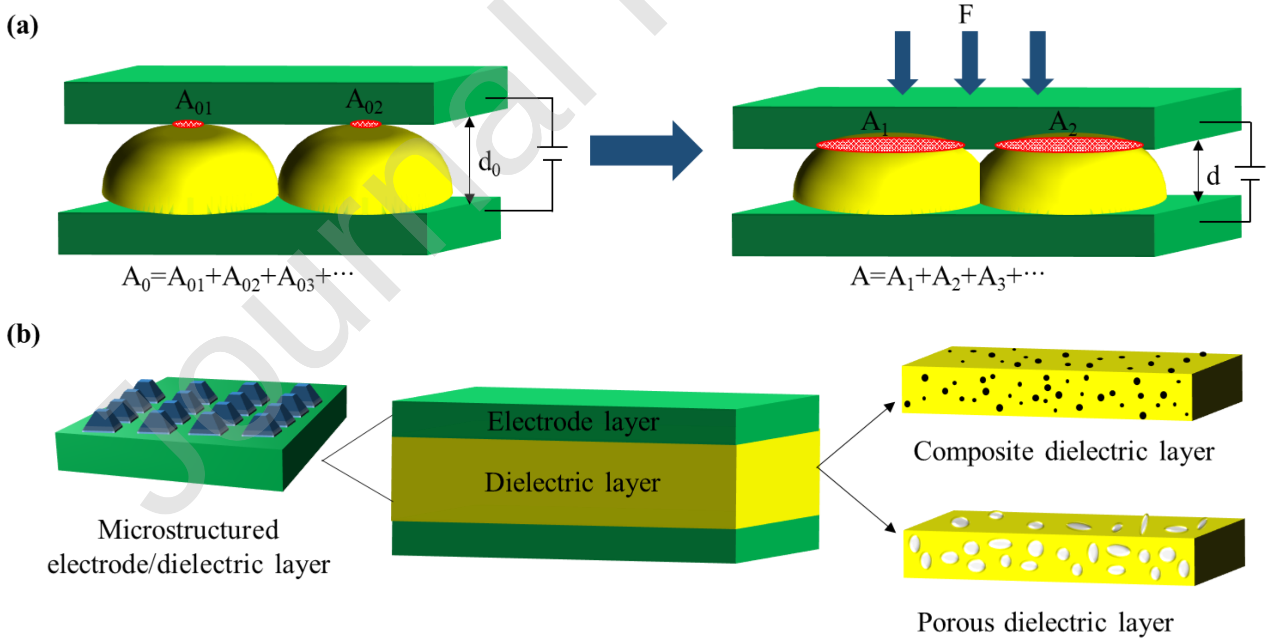


Fig. 3. Schematic diagram of, (a) variation of distance or area of electrodes of capacitive pressure sensor, (b) three effective ways to improve the sensitivity of capacitive sensor.

4. Improving sensitivity of flexible capacitive pressure sensor

4.1 Classification of improvement methods

Sensitivity is an important parameter to decide a sensor performance. It is usually defined by the ratio of the sensor output to the input signals, reflecting the accuracy and effectiveness of a sensor in practical [4]. The sensitivity of a capacitive pressure sensor is defined as:

$$S = \frac{(C/C_0)-1}{\Delta P} \qquad (2)$$

By combining the capacitance equation, the sensitivity can be calculated by Eq. (3):

$$S = \frac{\left(\frac{A}{A_0}\times\frac{d_0}{d}\times\frac{\varepsilon_r}{\varepsilon_{r0}}\right)-1}{\Delta P} \qquad (3)$$

Where S represents the sensitivity, C_0 is the initial capacitance of the capacitive pressure sensor, C is the capacitance after applying a load, ΔC is the capacitance variation, ΔP is the pressure variation, A_0 and A, d_0 and d, and ε_{r0} and ε_r represent the relative area, distance and relative dielectric constant of two layers of electrodes before and after applying loading. The greater the S value is, the easier the sensor can sense the pressure change. It can be seen from Eq. (3) that, under the same loading, any way to improve the sensitivity requires a change of the value of A, d or ε separately or simultaneously.

The Young's modulus of some commonly used dielectric elastomer are high, and the distance of electrodes of a capacitive flexible pressure sensor is relatively hard to change under little pressure, resulting in a small capacitance variation and thus a low sensitivity. Until now, many studies have been attempted to improve the sensitivity in order to extend the applications of capacitive flexible pressure sensors. After review, it is concluded that the effective ways to improve the sensitivity of sensor mainly have three aspects, as shown in Fig. 3(b): a) to construct microstructure of the dielectric or electrode; b) to add conductive fillers to polymer elastomer to generate a composite dielectric; c) to introduce micro-holes into the dielectric layer.

4.2 Research advances of improvement ways

(1) Microstructured dielectrics/electrodes

Construction of microstructures in dielectric or electrode has been one main research way in improvement of the sensitivity of flexible capacitive pressure sensor. To have microstructures for dielectric elastomer/electrodes gives rise to a decrease of their viscoelasticity that can avoid the increase of sensor hysteresis, and a relatively easier deformation of the microstructured dielectric/ electrode is observed due to a reduced Young's modulus [40]. Furthermore, to have microstructures would involve air into the sensor which is able to improve the effective dielectric constant to benefit the sensitivity When a load is applied to a micro-structured sensor, the variation ratio of capacitance would increase, giving rise to an improved sensitivity of sensor. Here, the effective dielectric constant of the sensor with microstructures can be estimated as:

$$\varepsilon_{eff} = \varepsilon_a V_a + \varepsilon_p V_p \tag{4}$$

Where ε_{eff} is the effective dielectric constant of the dielectric layer, ε_a、ε_p are the dielectric constant of air and the elastic dielectric material ($\varepsilon_p > \varepsilon_a$), respectively, V_a、V_p are the volume fraction of the air gap and the elastic dielectric material, respectively[12, 22, 41]. Pyo et al. [42] described that air is more easily compressed than common elastic dielectric materials. Therefore, V_a decreases with the increase of V_p when the pressure sensor is under loading. It can be calculated that ε_{eff} increases correspondingly from Eq. (4).

In 2010, Bao et al. [17] introduced the conception of microstructures into the dielectrics for the first time, providing a new method for improving the sensitivity of sensors. In the work, a PDMS dielectric layer was prepared with pyramidal microstructures by using silicon mould, the sensitivity of the sensor reached 0.55 kPa^{-1}, as shown the tested sensitivity in Fig. 4(a). After a comparison, it is found that the sensitivity of the micro-structured sensor is much higher than the sensors using line-structured film and the unstructured film. The study results provide a new innovative way to create the dielectric layer with low elastic modulus, which has attracted a later extensive attention and research in academia.

Later, many studies on microstructures were performed using photolithography, chemical etching and other methods. For example, Luo et al. [40] developed a dielectric layer with tilted microstructure. In the study, a template with tilted microstructure was obtained by tilted photolithography first. Then, a layer of PDMS was uniformly covered onto the mold surface by pouring and spin-coating. Before PDMS curing, it was bonded with Au/PET electrode layer and demolded together after curing. The top interface of the microstructure was in contact with the flexible PDMS. The interface of the microstructure pattern is in a bending state when subjected to a load, which is more prone to a deformation than compression. Fig. 4(b) presents the measured sensitivity of the sensor with the value of 0.42 kPa^{-1} in the low pressure range of 0-1 kPa, which is higher than many types of common sensors, as shown a comparison of the sensitivities of a few typical developed sensors in Table 1. It can be seen that to construct the microstructured dielectric layer is an effective way to improve the sensitivity. However, the generation of the microstructures usually leads to an increased thickness of dielectric. It was reported that the sensitivity of a thin dielectric layer is higher than that of the thick dielectric layer [53] using the same materials, thus except for the ease of deformation, the generation of microstructures on the dielectric layer will affect the improvement of the sensitivity to some extent.

Table 1

A comparison of sensitivities between [40] and other common sensors.

Dielectric material	Electrode	Sensitivity	Reference

Microstructured PDMS	Au/PET	0.42 kPa^{-1}	[40]
Ecoflex	Ag	0.0224 kPa^{-1}	[43]
Ecoflex	Ag/PET/PDMS	0.00145 kPa^{-1}	[44]
Very high bond (VHB)	Hydrogel	0.09 kPa^{-1}	[45]
Ecoflex	AgNW/PDMS	0.00162 kPa^{-1}	[46]
Ecoflex	poly(3,4-ethylenedioxythiophene): poly(styrenesulfonate) (PEDOT:PSS)	0.0077 kPa^{-1}	[47]
Ecoflex	Carbon nanotubes (CNTs)/PDMS	0.0004 kPa^{-1}	[48]
PDMS	Au/PDMS	0.0036 kPa^{-1}	[49]
PDMS	Cu/PI	0.04 kPa^{-1}	[50]
Ecoflex	CNTs/PDMS	0.0002 kPa^{-1}	[51]
PDMS	Au/PI	0.0018 kPa^{-1}	[52]

Besides the dielectric layer, Li et al. [38] proposed that the introduction of microstructures onto the electrode was also an effective way to improve the sensitivity. In the work, a micro-pyramidal PDMS was obtained by using a template, and then deposited Au on its surface to act as a microstructured electrode. A sensor was fabricated using the microstructured electrode and an ultra-thin dielectric layer. In order to avoid the viscoelasticity of the thin dielectric layer, parylene was selected as the dielectric material. As shown in Fig. 4(c), the sensor shows the measured ultra-high sensitivity of 70.6 kPa^{-1}, which is a much higher value than the value of the sensor using the same microstructured dielectric layer. The sensor using microstructured electrodes shows great application prospects in the areas of e-skin, medical monitoring devices, etc.

Besides the pyramidal microstructures, there are many kinds of microstructures that have been developed. For example, Cheng et al. [54] developed a kind of hierarchical pyramidal microstructures. The inverted pyramidal structures with various sizes were formed on the surface of a silicon mould by photolithography and wet etching, and then a PDMS layer was released from the mold and bonded with Pt element to obtain a hierarchical microstructured electrode, as shown in Fig. 4(e). The hierarchical structure has reduced the density of the large pyramids, thereby improving the sensitivity of sensor. Meanwhile, an insertion of small pyramids can reduce the hysteresis of sensor, avoiding some influences on sensitivity. It was also found that the sensor using hierarchical microstructured electrodes has the characteristics of low hysteresis and high sensitivity (up to 3.73 kPa^{-1}, higher than 0.66 kPa^{-1} in Ref. [17], 0.21 kPa^{-1} in Ref. [16], and 1.43 kPa^{-1} in Ref. [55]). The sensor can realize the accurate acquisition of pulse. Yang et al. [56] prepared an electrode with a microstructured structure different from that of a work [54] by photolithography. In the work, a copper mould with a dome was obtained by photolithography and wet etching. After the graphene was evenly distributed in the mould, it was bonded with PET to obtain a microstructured electrode. When a single layer of microstructured electrode is used for a sensor, the sensitivity was measured to be 3.19 kPa^{-1}. This sensor can be used for pulse monitoring and object grasping, and has great application potential in areas of robot control and human-machine interaction. If a double layer of symmetrical microstructued electrodes was used for a sensor, the sensitivity was measured to be 7.68 kPa^{-1}, as shown in Fig. 4(d). This indicates that the microstructured electrode plays an important role in improving the sensitivity of sensor.

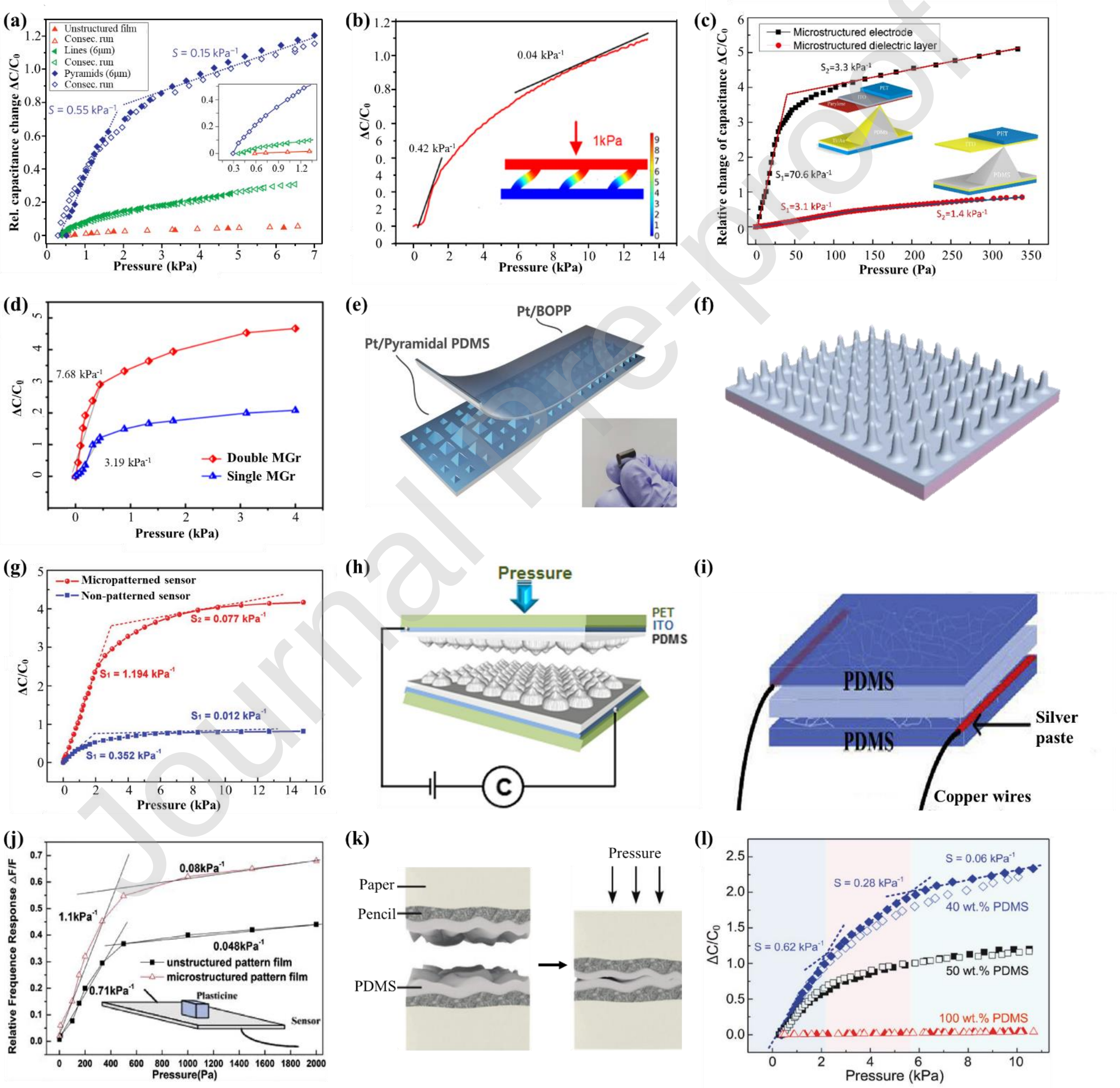


Fig. 4. Measured results of, (a) pressure-response curves for sensors with different fabricated structures, (b) pressure-response curve for tilted microstructure dielectric layer, (c) pressure-response curves for a SMD (sensor with microstructured dielectric layer) and a SME (sensor with microstructured electrode) with the same pyramid dimensions, (d) relative capacitance curve for two kinds of pressure sensors with single micro-structured graphene electrode and double micro-structured graphene electrodes against increasing pressure; schematic diagram of, (e) the hierarchically structured pressure sensor, the inset shows an optical image of the sensor, (f) the PDMS flexible substrate with lotus leaf microtower structure, (g) sensitivity of the micropatterned and nonpatterned sensor, (h) the capacitive pressure sensor based on the rose petal microstructure, (i) the capacitive pressure sensor with microstructured PDMS/AgNWs electrode film, (j) sensitivity of sensor with unstructured pattern film and sensor with microstructured pattern film, (k) the paper-based pressure sensor under loading, (l) relationship curves of relative capacitance change and pressure of the flexible pressure sensors with different dielectric layers .

Although the processes of photolithography, chemical etching, wet etching and other methods can produce high-precision and high-resolution microstructures [24], they are still not suitable for industrial production due to the high cost. In view of this problem, some studies employed natural materials as templates for the low cost and common processing ways. For example, Su et al. [57] made a great progress in improving the sensitivity of piezoresistive sensor by copying mimosa leaves. Inspired by this, some natural materials were also used as templates for making capacitive pressure sensors. For example, Wan et al. [58] took lotus leaf as template, and obtained PDMS flexible substrate with lotus leaf microtower structure, as shown in Fig. 4(f). In order to ensure the microstructures were highlighted in sensor, an ultra-thin AgNWs film was coated on the surface of the microstructured PDMS to obtain a flexible electrode layer. A capacitive pressure sensor with a high sensitivity was obtained by assembling the microstructured flexible electrode, the colorless polyimide (CPI) dielectric layer and the AgNWs/PDMS upper electrode. In the pressure test range of 0-2 kPa, the sensitivity of the sensor was measured to be 1.194 kPa^{-1}, as shown in Fig. 4(g). This is a much higher sensitivity than that of the non-patterned sensor. The high-sensitivity sensor can be used to detect the touching sense, heartbeat and airflow, and has broad application prospects in many areas.

In nature, there are also many biological surfaces with microstructures in addition to the lotus leaf. Mahata et al. [59] covered polymethylmethacrylate (PMMA) on the rose petal and obtained a PMMA template with the inverted micropapillae structure. The PDMS solution was poured onto the PMMA template, and then released after curing to obtain the microstructured PDMS dielectric layer with the same structure as the rose petal. After lamination, the microstructured PDMS dielectric layer was combined with the upper and bottom indium tin oxide (ITO)/PET electrodes to obtain a pressure sensor based on the rose petal microstructure, as shown in Fig. 4(h). The results showed that the sensitivity of the sensor is 0.055 kPa^{-1}, which is superior to some reported devices such as a thin all-elastomeric capacitive pressure sensor (4×10^{-4} kPa^{-1}) reported in Ref. [48], poly (glycerol sebacate) elastic dielectric based capacitive pressure sensor (0.11 kPa^{-1}) fabricated in Ref. [60] and polymer dielectric film with a nano-needle structure based capacitive pressure sensor (0.0268 kPa^{-1}) developed in Ref. [61]. The reason for the high sensitivity of the sensor is that the hierarchical micropapillae structure is relatively easy to deform under pressure. The contact area increases under a load, then the distance between electrodes decreases, and the dielectric constant increases due to the air discharge. It has been proved that the sensor can be used to monitor the radial artery pulse, blood pressure and heart rate, and it has indicated a certain application value in physiological signal monitoring.

In daily life, there are many objects with microstructures on the surfaces. Quan et al. [62] developed a highly sensitive flexible pressure sensor with a microstructured PDMS/AgNWs electrode film using matte surface glass as template, as displayed in Fig. 4(i). The sensor employed AgNWs as a kind of conductive material that was deposited on the surface of a microstructured PDMS substrate to form an electrode layer. Here, the flat PDMS film acts as the dielectric. In the pressure range of 0-500 Pa, the sensitivity of the sensor was measured to be 1.1 kPa^{-1}. The value is higher than 0.71 kPa^{-1} of the sensor with the smooth flat glass template, as presented in Fig. 4(j). The sensor was reported to have a potential use in sensing the pressure of mouse click, and may have a good application prospect in flexible touch screen and health care. In another study, as shown in Fig. 4(k), a flexible paper was used to fabricate the capacitive flexible pressure sensor. The rough surface (in similarity with the microstructure) of the paper was used to improve the sensitivity of the sensor [63]. A graphite conductive layer was transferred to the paper by pen writing. This transfer method maintains the rough surface of the paper as a microstructured electrode layer. Then, the PDMS solution diluted with heptane (40wt% PDMS) was coated thinly on the surface of the electrode layer as the dielectric layer, and finally a capacitive flexible pressure sensor was assembled using another same half structure. The results showed the sensitivity to be 0.62 kPa^{-1}, which is higher than the sensitivity of the sensor with flat surface (100wt% PDMS), as shown in Fig. 4(l). It was reported that using the rough surface of paper is a simple and economical way to improve the sensitivity of sensor. However, there are also some issues like low precision and difficult to control that limits its further application.

Here, using natural materials is convenient and environmental friendly, however, the size of the inherent microstructure is hard to control to ensure uniformity. Therefore, some researchers use relatively cheap templates or other economic approaches to fabricate microstructures. Guo et al. [22] fabricated a highly sensitive pressure sensor with a double-sided microstructured dielectric layer using commercial anodized aluminum oxide (AAO) as template. Poly(vinylidenefluoride-co-trifluoroethylene) (P(VDF-TrFE)) with high dielectric constant and low viscoelasticity was used as the dielectric material. The dielectric layer with double-sided nanopillars was obtained by hot pressing using two identical AAO templates, and then it was assembled with

Cu/PI electrodes to obtain a sensor as shown in Fig. 5(a). The sensitivity of the sensor was measured to be 0.35 kPa^{-1}, which is superior to the unstructured sensor of 0.12 kPa^{-1}. As shown in Fig. 5(b) and (c), the sensor has been used for monitoring human heartbeat, pulse, breathing and speech recognition, the related signals were successfully obtained, indicating the strong feasibility of such materials in developing flexible pressure sensors. In another work, the same materials were selected to make sensors, and a new interlocked microstructure was used to obtain a flexible pressure sensor with higher sensitivity. Inspired by the interlocked structure of human skin, Niu et al. [41] used the AAO template to fabricate the upper and bottom P(VDF-TrFE) dielectric layers with the interlocked asymmetric nanocones structure, as illustrated in Fig. 5(d). For an applied pressure in the range of 0-100 Pa, the sensitivity of the sensor was measured to be 6.583 kPa^{-1}, which is higher than that of work [22]. This is ascribed to the microstructure used in the study is easier to deform under pressure, and the variation range of d and ε are larger, resulting in a higher sensitivity.

In other studies, different moulds or methods were used to fabricate microstructured sensors. Joo et al. [31] prepared a highly sensitive flexible pressure sensor with microstructured electrode based on a pre-strained PDMS template. The pre-strained PDMS was treated with ultraviolet/ozone (UV/O_3) to form a layer of silicon oxide (SiO_X) thin film on the surface, and then coated with a layer of AgNW film. When a tension applied on the PDMS was removed, the PDMS mould with bending structure and the AgNW electrode film was obtained, then, a PDMS solution was completely covered on the surface. After curing, the microstructured AgNW/PDMS electrode layer was obtained by releasing together with the AgNW electrode film as shown in Fig. 5(e). The wavy structure of the electrode and the rough surface formed by AgNW on the microstructure endows the capacitance of the sensor with a great variation, exhibiting a higher sensitivity (3.8 kPa^{-1}) than that of the sensor with non-structured electrode (~0). The experiment verified that the sensor has an ability to monitor the pressure of fingertip and has a potential use in the muscle rehabilitation training equipment.

Zeng et al. [64] utilized the same method as the work [31] to manufacture the PDMS moulds. The morphology of the microstructure was adjusted by changing the tension and the ultraviolet ozone (UVO) processing time during the manufacturing. Some holes were introduced into the microstructure by changing the curing time of PDMS and applied pressure during curing. Under the joint action of these aspects, a pressure sensor with high sensitivity was obtained as shown in Fig. 5(f). Fig. 5(g) shows that the sensitivity of the microstructured sensor is much higher than that of the unstructured one. The sensitivity of the microstructured sensor was measured to be 14.268 kPa^{-1} in the pressure range of less than 0.7 kPa^{-1}, and 0.032 kPa^{-1} in the pressure range of 0.7-40 kPa^{-1}. The sensor can be used for speech recognition and breathing rate monitoring. In another study, the pre-stretched PDMS was etched by plasma and then released to obtain a PDMS microstructured template [37]. The microstructured PDMS dielectric layer was then tightly bonded with the bottom electrode layer of AgNMs/PDMS, and assembled with the upper electrode layer of AgNMs/PDMS to fabricate a flexible pressure sensor. The microstructured dielectric layer endows the sensor with a high sensitivity. As shown in Fig. 5(h), the sensitivity was 2.04±0.16 kPa^{-1} in the low pressure range of 0-2 kPa, and 0.57±0.08 kPa^{-1} when the pressure rises to the range of 2 kPa-9 kPa, which is much higher than the pressure sensor using flat dielectric layer.

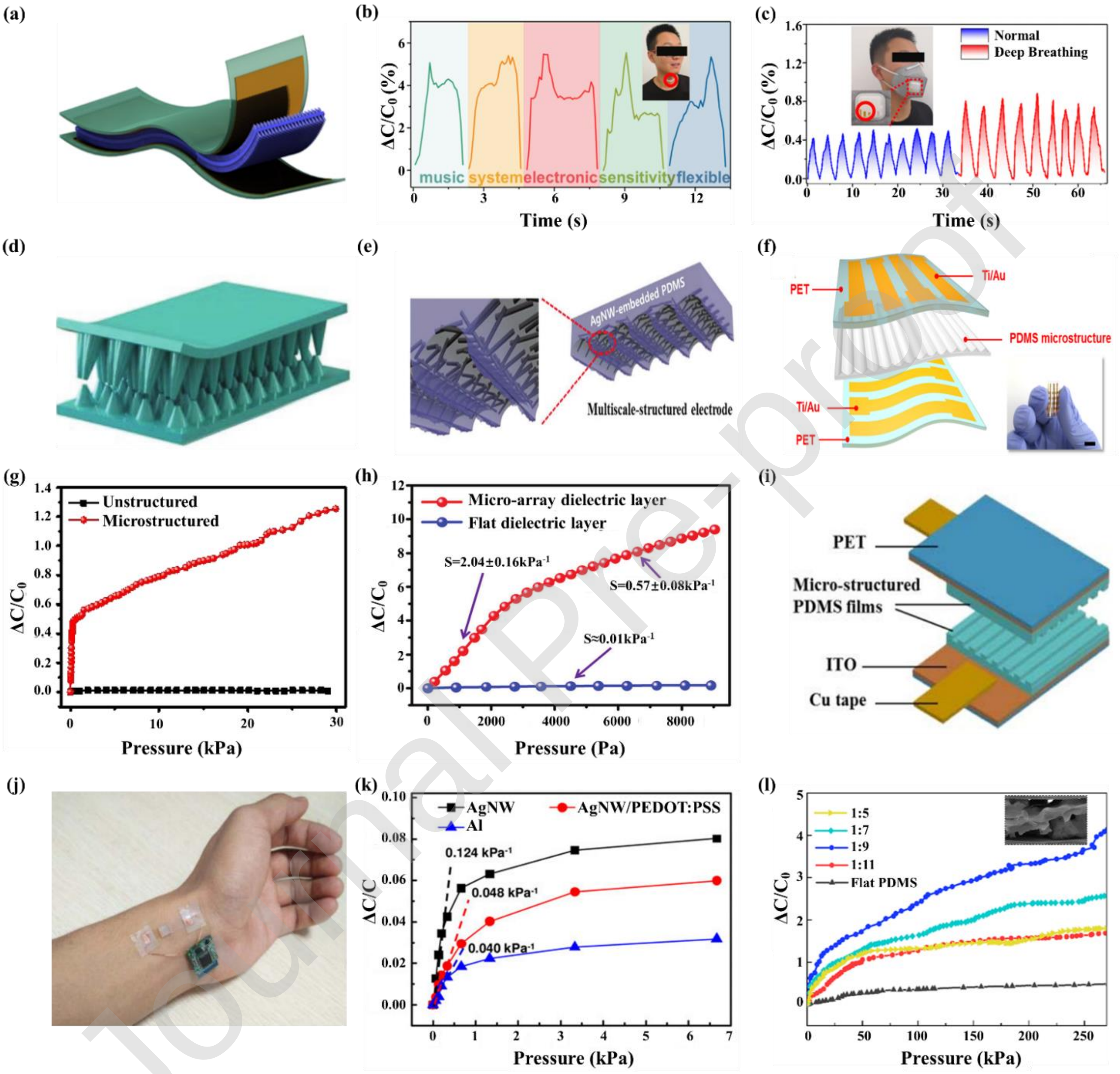


Fig. 5. Methods of fabricating microstructures, (a) schematic diagram of a pressure sensor with a double-sided microstructured dielectric, capacitance response curves, (b) when pronouncing different words and (c) when breathing, (d) the dielectric layer with an interlocked asymmetric nanocones structure, (e) schematic diagram of the microstructured AgNW/PDMS electrode layer, (f) schematic structure of the pressure sensor, the inset shows the flexibility of the sensor, (g) sensitivity of the microstructured and unstructured sensors, (h) relative capacitance change-pressure curves of the flexible pressure sensors with different dielectric layers, (i) illustration of the device structure of the fabricated capacitive pressure sensor, (j) photographic of wrist pulse measurement, (k) sensitivity of

sensors with different electrode materials, (l) relative capacitance and change-pressure curves of the capacitive pressure sensors with different dielectric layers, the inset shows the SEM image of the microstructured dielectric layer when the ratio of instant soluble sugar to salt is 1:9.

3D printing is a rapid prototyping method applied in many areas in recent years, for example, mould printing would gradually replace a few complex processes such as photolithography and wet etching. In fabrication of flexible pressure sensor, Zhuo et al. [65] used 3D printing to fabricate an acrylonitrile butadiene styrene (ABS) resin template with a covered PDMS solution to prepare a microstructured PDMS dielectric layer, and then combined with the ITO/PET electrode layer to obtain a flexible pressure sensor as shown in Fig. 5(i). The microstructured PDMS dielectric layer enables the sensor's sensitivity to be 1.62 kPa^{-1}, and it's higher than some previous works using silicon moulds prepared by micro-fabrication processes such as 2×10^{-4} kPa^{-1} in Ref. [51], and 0.55 kPa^{-1} in Ref. [17]. The sensor can monitor human physiological weak signals such as human pulse, as shown in Fig. 5(j).

However, the manufacturing process of microstructures via commercial mould is complicated, and the microstructures obtained by copying biological surfaces are uncontrollable. For this issue, Chen et al. [30] proposed a method to obtain the microstructure without a template. By laminating two PDMS films embedded with AgNWs together, a highly sensitive capacitive flexible pressure sensor was prepared. The results showed that the rough structure formed by AgNWs on the PDMS surface plays a positive role in improving the sensitivity of sensor. From Fig. 5(k), it is noted that the rough surface of the AgNWs mesh electrode sensor presents a much higher sensitivity than the flat surface of the AgNW/PEDOT:PSS and Al film electrode devices. The rough surface increases the number of pores in sensor, leading it to deform easier under a pressure, and the sensitivity of the sensor reaches the value of 0.124 kPa^{-1}. Shao et al. [66] chose an amount of multi-walled CNTs (MWNTs) embedded in PDMS as the flexible substrate. Meanwhile, in order to improve the conductivity of electrode, a layer of Au film was deposited on the MWNTs film. The sensor with the MWNTs/Au electrode showed a higher sensitivity (1.33 kPa^{-1}) in comparison with some previous related works using various electrode materials, such as Au electrode (3.6×10^{-3} kPa^{-1}) studied in Ref. [67], Cu electrode (1.8×10^{-3} kPa^{-1}) prepared in Ref. [52] and Al electrode (4.0×10^{-2} kPa^{-1}) reported in Ref. [50], respectively. Inspired from the traditional Japanese food Komochi konbu, Wang et al. [68] used the instant soluble sugar and salt as sacrificial materials to form inwardly curling convex and some internal micropores on the upper and lower sides of PDMS dielectric layer, as shown the inset of Fig. 5(l). The structure and the morphology of dielectric layer can be controlled by the ratio of instant soluble sugar to salt due to the difference in dissolution rate of the sacrificial materials. When the amount ratio of instant soluble sugar to salt is 1:9, the sensor with microstructured dielectric layer shows the sensitivity of 0.171 kPa^{-1}, a much higher value than that of a flat PDMS sensor (8.35×10^{-3} kPa^{-1}), as shown in Fig. 5(l). Moreover, the work of [30], [66] and [68] provided methods for obtaining microstructures without templates, but the sensitivity of the sensors is not as high as that of the sensors using templates.

(2) Composite dielectrics

To date, the composite dielectric has been widely used in piezoresistive sensors. However, such dielectric is still limited use in capacitive flexible pressure sensors. The study of composite dielectric in improving sensitivity of capacitive sensors is relatively late in comparison with the study of microstructure. A composite dielectric is usually formed by adding an amount of conductive fillers or ceramic fillers with high dielectric constant to an elastic dielectric material. The selected kinds of conductive fillers are similar to the conductive materials for electrodes, mainly including carbon nanotubes, carbon black, graphene and other carbon materials and metal particles, metal nanowires and other metal materials. In addition to the conductive fillers, some ceramic particles with high dielectric constant are used as fillers. For the polymer filled with conductive fillers, its conductivity increases sharply and varies from insulator to conductor when the filler content reaches a certain threshold. This refers to the percolation threshold [69]. The dielectric constant of the composite dielectric layer increases with the increase of the content of filler if it is lower than the percolation threshold. Then, it reaches the maximum value at the percolation threshold [70]. Percolation is a common phenomenon in particle filled polymer composites. The percolation theory points out that the probability of particles agglomeration increases with the increase of conductive particles content [71]. The particles in the polymer form a conductive path when the content of particles reaches the percolation threshold, resulting in the polymer to be conductive suddenly [71, 72]. The relationship between the dielectric constant of the composite dielectric and the filler content can be predicted by the percolation theory formula [73, 74]:

$$\varepsilon_r \propto (P_c - P)^{-s}, P < P_c \qquad (5)$$

Where ε is the dielectric constant of the composite dielectric layer, P_c is the percolation threshold of the filler, P is the filler content, and s is the critical exponent of dielectric constant which is about 0.64. It is noted from Eq. (5) that the dielectric constant of the composite is directly proportional to the filler content when the content is lower than the percolation threshold. However, the relationship is inversely proportional when the content is higher than the percolation threshold [75]. Therefore, the conductive filler content should be controlled within the percolation threshold to achieve the best result.

The sensitivity of the composite dielectric layer is mainly agitated by changing the dielectric constant which is closely related to the percolation threshold of composite dielectric layer [29, 76]. However, the percolation threshold of composite dielectric layer varies with the applied pressure [77, 78]. Under a pressure, the distance of conductive particles changes, which will reduce the percolation threshold [79, 80]. According to Eq. (5), P_c decreases with the increase of ε_r. Moreover, the relative capacitance change can be calculated by [81]:

$$\frac{\Delta C}{C_0}=\frac{C-C_0}{C}=\frac{\varepsilon_0\varepsilon_r(A/d)}{\varepsilon_0\varepsilon_{ro}(A/d_0)}-1=\frac{\varepsilon_r}{\varepsilon_{r0}}\cdot\frac{d_0}{d}-1 \qquad (6)$$

Where ΔC represents the capacitance variation, C_0 is the initial capacitance, d_0 and d represent the distance between electrodes without and with loading, respectively, and ε_{r0} and ε_r represent the dielectric constant without and with loading, respectively. For the sensor based on composite dielectric, $\varepsilon_r>\varepsilon_{r0}$, while in the case of the sensor based on non-composite dielectric, $\varepsilon_r=\varepsilon_{r0}$. The ΔC of the sensor with composite dielectric layer becomes larger with the increase of dielectric constant, therefore, the sensitivity is improved according to the Eq. (2) [81]. It's concluded from Eq. (5) and Eq. (6) that the dielectric constant of dielectric layer is not affected by the concentration of fillers only, but also affected by the compression behavior.

Metal is a kind of common conductive material that can be used as candidate filler in dielectric. For example, nanoparticles like Ag, Cu, Au and other metals are often used as fillers, and the Ag nanoparticles and nanowires are the mostly studied conductive fillers. Liu et al. [82] fabricated a high sensitivity of the sensor by 101.5% by adding some Ag nanoparticles into the PDMS dielectric. Although the sensitivity of this sensor is still less than that of piezoresistive and piezoelectric sensors, the sensor to some extent can sense very tiny pressure changes from pulse, as shown in Fig. 6(a). The metal nanowires have a higher aspect ratio and are more suitable as fillers than the nanoparticles. Mi et al. [83] proved that a small amount of Ag nanowires (AgNWs) can significantly improve the dielectric constant of polyurethane (TPU) dielectric, giving rise to a widely use of AgNWs in composite dielectric of sensors. Wang et al. [81] mixed AgNWs and PU aqueous solution for a mixed conductive ink, which can be spun-coated on an electrode surface, then assembled with the other half for fabrication of a pressure sensor. With an increase of concentration of AgNWs filler from 0 to 3wt%, the physical distance between fillers was reduced, the dielectric constant was then increased from 5.4 to 45.3, and the constant would be more if the dielectric was under a load, as shown in Fig. 6(b). Here, the sensitivity of the sensor was measured to be 5.54 kPa^{-1} when the pressure of a load was less than 30 Pa, while that of the sensor with pure PU dielectric was 0.50 kPa^{-1}. Furthermore, when the pressure was more than 30 Pa, the sensitivity of the two sensors was 0.88 kPa^{-1} and 0.23 kPa^{-1}, respectively. After verification, the sensor can be used to detect body and muscle movements, and even some non-contact pressures that are similar to Fig. 6(c). This indicates a great application potential in airflow detection.

Owing to the high cost of Ag nanomaterials, its application is still limited. In contrary, the low cost metal Zn is attractive to be used as fillers. Chen et al. [84] added ZnO nanowires into PMMA solution and spin-coated them uniformly on the surface of electrode to obtain a composite dielectric layer, as shown the schematic diagram of the sensor in Fig. 6(d). In addition to the change of distance of electrode layers, the separation of Zn^+ and O^- also increases the capacitance change. The sensitivity of the sensor was measured to be 9.95 kPa^{-1}, which is 23 times higher than that of pure PMMA dielectric layer sensor (0.43 kPa^{-1}).

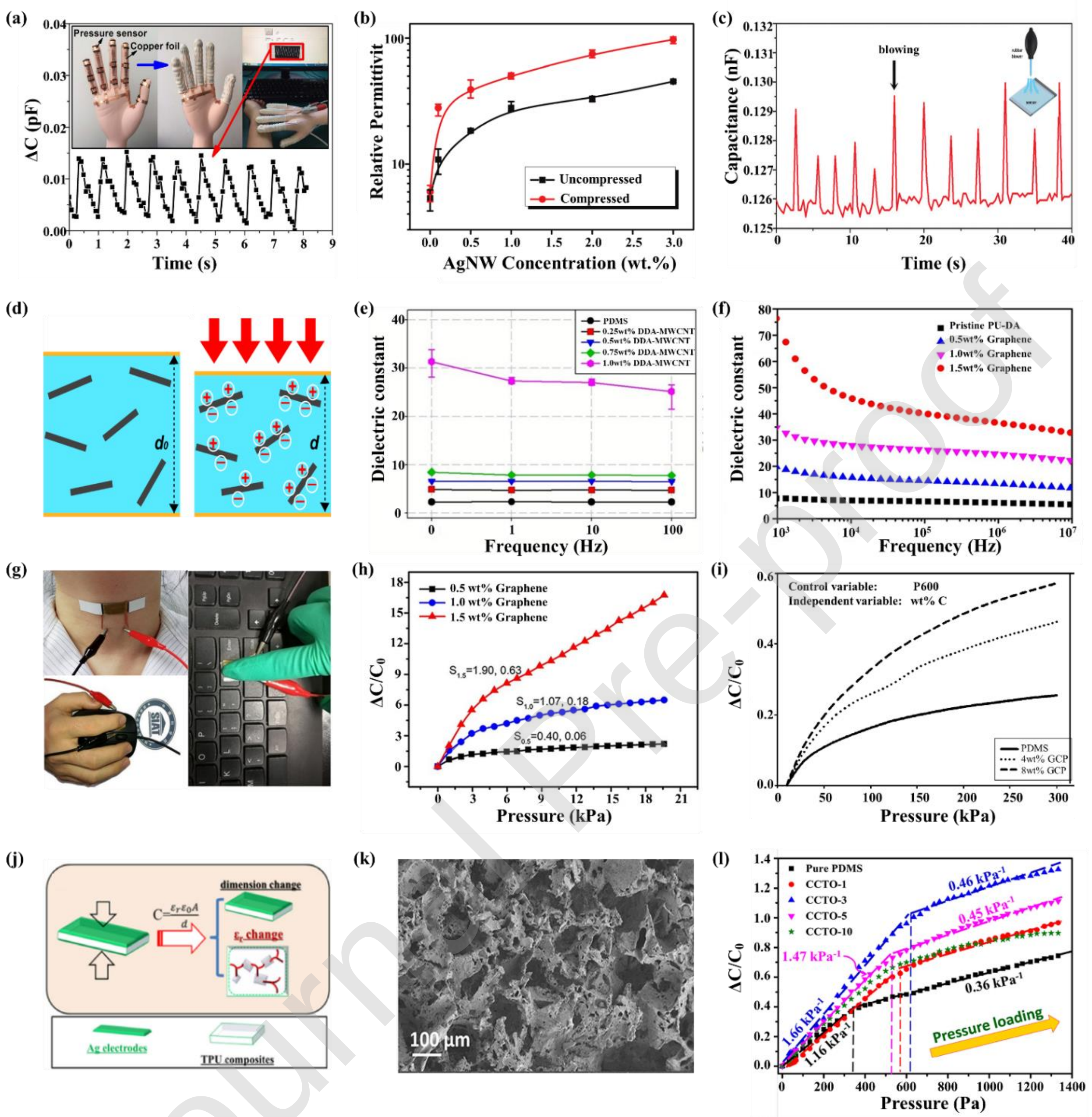


Fig. 6. Sensors with composite dielectrics with the measured results, (a) capacitance response curve obtained by touching the wrist pulse with an artificial hand, (b) at 1.1 GHz, relative dielectric constant curves of compressed and uncompressed dielectric layers at different AgNW concentrations, (c) schematic diagram of non-contact pressure detection and its corresponding capacitance signal peaks, (d) schematic diagram of the pressure sensor before and after compression, (e) dielectric constant of composites as a function of frequency, (f) dielectric constant versus frequency of dielectrics with 0.5, 1.0, and 1.5 wt% of graphene, (g) application of the pressure sensor in mouse click, keyboard click and speech recognition, (h) relative capacitance change-pressure curves of the capacitive pressure sensors with different dielectric layers (with 0.5wt%, 1.0wt% and 1.5wt% content of graphene), (i) output variation of the sensor with different carbon black content obtained by the control variable method, (j) capacitance change schematic diagram of the capacitive pressure sensor composed of composite dielectric layer, (k)

SEM images of $CaCu_3Ti_4O_{12}$-PDMS (CCTO-PDMS) dielectric layer under different magnification for CCTO content of 5wt%, (l) relative capacitance variation–pressure curves of the capacitive pressure sensors with different dielectrics (with 0%, 1wt%, 3wt%, 5wt% and 10wt% content of CCTO).

According to the percolation theory, by adding conductive materials into an insulating polymer dielectric below its percolation threshold can change the electrical properties of the dielectric significantly. This can improve the relative dielectric constant [71], and then achieve the purpose of improving the capacitance change. Carbon materials have good chemical stability and conductivity. A small amount of carbon materials can significantly improve the dielectric constant [85], which are another most commonly used fillers besides metal nanomaterials. Then, Jang et al. [86] added alkylamine modified multiwalled carbon nanotubes (MWCNTs) into PDMS elastomer to form a composite dielectric. From Fig. 6(e), it can be seen that the dielectric constant was increased greatly by adding MWCNTs into the dielectric. Only 1.0wt% alkylamine modified MWCNTs in pure PDMS dielectric layer would increase the dielectric constant from 2.8 to 8.0, while the capacitance was increased by 1.8 times, and the sensitivity was improved greatly either. CNTs as fillers have been often studied together with other methods which will be reviewed in section (4).

Graphene as conductive filler is another commonly used carbon material in fabrication of sensor. Liu et al. [87] distributed graphene into healable polyurethane (HPU) solution uniformly by ultrasonic agitation to improve the dielectric constant of dielectric. From Fig. 6(f), it is noted that an addition of graphene can change the dielectric constant of HPU significantly. Meanwhile, as the content of graphene gradually increases within the percolation threshold, the sensor sensitivity also increases, as shown in Fig. 6(h). When the graphene content is 1.5wt%, the sensitivity of the sensor is increased to 1.9 kPa^{-1} (<3 kPa), which can be used for detecting fingertip pressure and vocal cord vibration, as shown in Fig. 6(g). The addition of carbon nanotubes and graphene fillers can improve the relative dielectric constant and optimize the sensitivity of pressure sensor greatly, however, these filler materials are expensive and not suitable for mass production. Owing to this reason, Ma et al. [26] chose carbon black as filler for its low cost and good conductivity. The carbon black was mixed with PDMS to prepare the composite dielectric. The performance of the sensor was greatly improved by controlling the content of carbon black, as illustrated in Fig. 6(i).

A combination of conductive filler with polymer elastomer causes some loss of dielectric, which affects the sensor sensitivity more or less. Here, Ke et al. [88] prepared a sensor with adjustable dielectric constant. In the sensor, a newly developed high conductivity branched carbon nanotubes (CNS) and the graphene nanoplatelets (GNP) with high dielectric constant and low dielectric loss were used to increase the dielectric constant of the TPU elastomer and restrain its dielectric loss. The relative dielectric constant of the composite dielectric can be changed by adjusting the content of CNS and GNP. Under the filling conditions, 1.5wt% CNS, 0.5wt% GNP and 1 kHz, the dielectric constant of the composite dielectric reaches 2400, i.e., 300 times higher that of pure TPU dielectric.

As shown in Fig. 6(j), the distance decreases and the relative area increases for the two electrodes under pressure. Meanwhile, the dielectric constant increases due to the effect of conductive fillers, and the prepared sensor sensitivity is thereby as high as 2.05± 0.13 MPa^{-1}, which is significantly higher than 0.18± 0.01 MPa^{-1} of the sensor with pure TPU dielectric.

Apart from using conductive fillers to increase dielectric constant, some people attempt ceramic nanoparticles with high dielectric constant to improve the dielectric constant of dielectric layer. For example, Mu et al [89] added $CaCu_3Ti_4O_{12}$ (CCTO) ceramic nanoparticles together with NaCl into PDMS solution. A porous PDMS dielectric layer containing CCTO ceramic nanoparticles was obtained by removing NaCl as shown in Fig. 6(k), while the ceramic nanoparticles can make up for the small dielectric constant of porous layer. As can be seen from Fig. 6(l), the sensitivity of the sensor was measured to be 1.66 kPa^{-1}, which is higher than the value of pure PDMS-based sensor (1.16 kPa^{-1}). This indicates it can be used for detection of radial pulse and Morse code. Rana et al. [90] also used a way to increase the dielectric constant by adding nanoparticles of ferroelectric ceramic to the dielectric layer. By adding barium carbonate ($BaTiO_3$) nanoparticles with the dielectric constant of 4000 to the silicone elastomer, the comprehensive dielectric constant of the sensor was significantly improved，and the sensitivity was optimized.

In fact, the addition of nanofillers will lead to a higher modulus of the dielectric layer, and affect the sensitivity of sensor. However, it can be seen from many studies that the positive effect of the increase of dielectric constant is more attractive even a negative effect on mechanical properties, and the sensitivity of the sensor is finally improved.

(3) Porous dielectric layer

To fabricate dielectric layer with pores can reduce its Young's modulus. The distance of electrodes is decreased under an increased pressure, then the pores are gradually closed and the dielectric constant is also increased. Therefore, the capacitance change and the sensitivity are both increased, as illustrated in Fig. 7(a). There are many ways to generate the porous dielectric layer, and the most commonly used way is the sacrificial solvent method, that is, adding an intermediate material to the polymer elastomer as the pore making material, and then removing the intermediate material under specific conditions to obtain the porous elastomer.

Kwon et al. [33] put Ecoflex into a sugar cube mould for curing, and then dissolved the sugar cube with water to obtain a porous Ecoflex elastomer dielectric. The sensitivity of the obtained sensor by combining the porous elastomer with CNTs-elastomer electrodes is shown in Fig. 7(b). In the low pressure range (<5 kPa), the sensor sensitivity was measured to be 0.601 kPa^{-1}, which was used for real-time detection of wrist pulse successfully. Chen et al. [91] added ammonium bicarbonate (NH_4HCO_3), a common foaming agent, to PDMS solution. A porous PDMS dielectric was prepared after removing the sacrificial solvent by heating. The study provides a new method for the mass production of porous dielectric layers at low cost. The capacitive sensor was assembled using the porous PDMS dielectric layer and ITO/PET electrodes. Compared with the sensor using unstructured PDMS dielectric, the sensitivity has been significantly improved, as shown in Fig. 7(c). The sensor has high sensitivity in measuring a wide range of pressures, and can be used for real-time and reliable monitoring of pulse and plantar pressures, and thus has great application potential in human motion detecting.

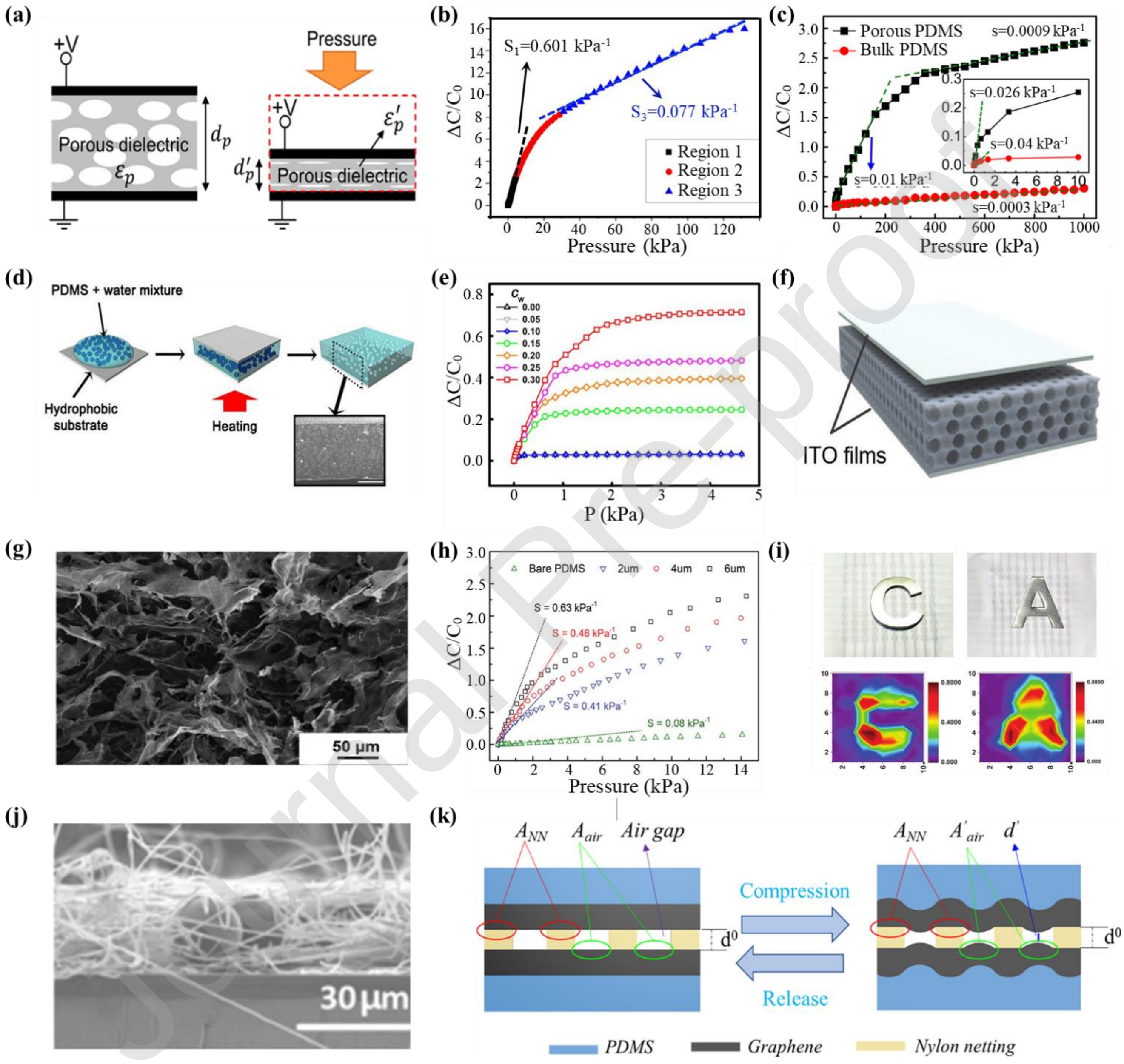


Fig. 7. Sensors with porous dielectric layers with measured results, (a) schematic of a porous capacitive pressure sensor under original state and it under a pressure state [23], (b) sensitivity of the sensor with a porous Ecoflex elastomer dielectric in different pressure ranges, (c) relative capacitance change-pressure curves of sensor with porous PDMS film and sensor with bulk PDMS film, the inset shows the relative capacitance change-pressure curves in low pressure (1-10 kPa) range, (d) preparation process of a porous PDMS film, (e) relative capacitance variation–pressure curves of the capacitive pressure sensors with different dielectrics (with 0.0%, 0.05%, 0.10%,

0.15%, 0.20%, 0.25% and 0.30% weight ratio of deionized water), (f) schematic of the sensor with uniform porous PDMS film prepared by polystyrene microspheres, (g) SEM images of Graphene oxide (GO) foam as dielectric layer, (h) sensitivity of three different porous structured pressure sensors and an unstructured pressure sensor, (i) sensing performance of the pressure sensor array to different letters, (j) SEM cross section of the nanofiber membrane, (k) working principle diagram of the pressure sensor with nylon net dielectric layer.

Although the work [33, 91] have improved the sensor's sensitivity by introducing a certain amount of micro-pores, the sensitivity in the low-pressure range is still limited, which constrains its use in e-skin and other areas requiring high sensitivity in the low-pressure range. Then, Lee et al. [92] dispersed deionized water in PDMS by stirring evenly, and prepared a layer of porous PDMS dielectric by heating for 24 hours. The preparation process is presented in Fig. 7(d). The sensitivities are different among the sensors fabricated with different weight ratios of deionized water (c_w). As shown in Fig. 7(e), the sensor has the highest sensitivity with c_w=0.3, which is much higher than that of the sensor using flat PDMS dielectric (c_w=0). The sensor (c_w=0.3) has a high sensitivity of 1.18 kPa^{-1} in the pressure range of < 0.02 kPa, and shows a high sensitivity in the range of <5 kPa, indicating a suitable candidate for manufacturing electronic touch screen, high sensitivity e-skin and other devices.

For the above studies, the micropores were randomly distributed in elastomers and their size was difficult to control. Then, Kang et al. [93] were inspired by spongia officinalis, and imitated the internal structure to fabricate sensors. Firstly, some polystyrene microbeads with the same size were evenly stacked together as template, and then PDMS solution was fully coated on the polystyrene microbeads. After curing, the PDMS dielectric layer with uniform porous structure was obtained by etching as shown in Fig. 7(f). The size of the micro-pores of the dielectric was regulated by the size of polystyrene microbeads. As is shown in Fig. 7(h), the sensitivity of the sensor using the dielectric and ITO/PET electrodes is 0.63 kPa^{-1}, which is considerably higher than the value of 0.08 kPa^{-1} of unstructured sensor. This indicates it suitable for making tactile perception of e-skin. However, this method is in the same way of the sacrificial solvent method, which introduces other materials in the matrix to make micro-pores. The process is too complex and expensive to be mass production. Wan et al. [94] directly sacrificed the solvent of the dielectric materials, frozen the graphene oxide solution at -50 ℃, and then dried it in vacuum to obtain the Graphene oxide (GO) foam dielectric layer, as shown in Fig. 7(g). The sensor using the GO foam dielectric layer has a sensitivity of 0.8 kPa^{-1} for the low pressure range (0-1 kPa) , which is 2×10^3 times higher than that of a flat PDMS dielectric layer [95], and 1.6×10^3 times higher than that of polyolefin foam [96]. Therefore, it can be used to monitor the pressure from tiny objects such as the petals (0.24 Pa).

The sacrificial solvent method displays the features of low cost while complex production process and time consuming, thus, the mass production via this method is difficult. Yang et al. [97] proposed a simple and time-saving method to prepare a porous dielectric by electrospinning. In the study, a porous thermoplastic poly urethane NM (TPUNM) dielectric was obtained with a high porosity, and provided a large space for the sensor to deform under a load. In addition, an electrode made of poly(vinylidene fluoride) NM (PVDFNM)/AgNWs was prepared by electrospinning and screen printing, and then the dielectric and PVDFNM/AgNWs were assembled by ultrasonic bonding. The prepared sensor has a higher sensitivity of 4.2 kPa^{-1} in comparison with the previous works, such as 0.76 kPa^{-1} in Ref. [17], 0.815 kPa^{-1} in Ref. [28] and 3.8 kPa^{-1} in Ref. [31]. The sensor can reliably monitor human heart rate, and even more the pressure sensor array can sense the pressure distribution of different letters, as shown in Fig. 7(i), which can provide aids for the preparation of e-skin. Yang et al. [98] also employed electrospinning to prepare the dielectric layer. During electrospinning, in order to increase the change of the relative dielectric constant under pressure, an amount of carbon nanotubes was added into polyvinylidene fluoride (PVDF) spinning solution to manufacture a porous composite nanofiber film dielectric layer. The capacitive sensor was obtained by covering ITO/PET electrode layers on the upper and lower sides of the dielectric layer. The sensitivity of the sensor was measured to be 0.99 kPa^{-1}, a higher value than that of the sensor without CNT (0.09 kPa^{-1}). However, it still has a certain difference with the sensitivity of work [97]. This may be ascribed to the different electrode structure in the two studies. In the latest study on electrospinning, Jin et al. [99] added a certain amount of insulating microbeads to the PVDF spinning solution to fabricate a porous PVDF dielectric layer by electrospinning. SEM images of the dielectric layer are shown in Fig. 7(j). The insulating microbeads improved the porosity of the dielectric layer significantly. The sensor's sensitivity in the pressure range of 0-1 kPa can reach 1.12 kPa^{-1}, which is superior to the most previous studies, such as 0.55 kPa^{-1} of the sensor with pyramid PDMS dielectric [17], 0.601 kPa^{-1} of the sensor with sugar sponge [23] and 0.759 kPa^{-1} of the sensor with yeast sponge [100]. Thus, the sensor is useful in the detection of human carotid pulse and respiratory rate.

Compared with the electrospinning of the work [97] to improve the sensor sensitivity, He et al. [13] adopted a nylon netting layer with regular microporous structure as the dielectric. The method is more simple, convenient and low cost, and avoids the viscoelasticity of the polymer dielectric layer. Two layers of graphene/PDMS flexible electrodes were respectively placed on the upper and lower sides of the nylon netting to form a capacitive sensor. Under a pressure, the relative area of the sensor electrode is increased, while the distance of two electrodes is decreased, as shown in Fig. 7(k). In the study, the performance of the sensor can be further optimized by changing the mesh numbers of the nylon netting. The sensor with 300-mesh nylon net exhibits the sensitivity of 0.33 kPa^{-1} in the low voltage range, which is higher than that of the sensor with 100-mesh nylon net (0.12 kPa^{-1}) and the sensor with 200-mesh nylon net (0.16 kPa^{-1}). After verification, the sensor can sense human pulse and mouse clicks, and has great potentials in biomedical detection and some wearable devices.

(4) Other methods

All of the above methods can improve the sensitivity of sensor effectively, but most of them are used solely. Then, some researchers have proposed a combination of two or more ways to improve the sensitivity appropriately. For example, Kim et al. [55] prepared a porous PDMS dielectric via the sacrificial solvent method, and introduced microstructure on the porous PDMS dielectric by using the commercial frosted glass as template. The process has increased the sensor sensitivity from 0.813 kPa^{-1} to 1.43 kPa^{-1}, as shown in Fig. 8(a). Atalay et al. [12] used a conductive knitted fabric as electrode and a porous silicon elastomer as dielectric to fabricate a flexible pressure sensor. Fig. 8(b) is the cross-sectional view that was captured via SEM. The conductive fabric electrode is equivalent to a microstructured electrode with an increased gap between the electrodes and dielectric. Combined the conductive knitted electrode with the porous dielectric layer, it is noted that the sensitivity of the sensor has been improved and higher than that of the sensor with unstructured knit, as shown in Fig. 8(c).

In addition to the combination with porous elastomer, the microstructure can also be assembled with composite dielectric to improve the sensor sensitivity. Rana et al. [90] mixed $BaTiO_3$ and Ecoflex uniformly. As shown in Fig. 8(d), the mixed solution was poured on a template based on laser engraving to obtain a composite dielectric with double-stage microstructure. Here, the microstructure improves the sensor compressibility under pressure, and the ceramic nanoparticles greatly increase the dielectric constant. Under the synergy of two aspects, the output signal of the sensor is amplified and the sensitivity is improved significantly. Shi et al. [18] fabricated a sensor using AgNWs as the filler of microstructured dielectric. In the study, the sensitivities of three types of sensors, that are, non-patterned without AgNWs, microstructure without AgNWs, microstructure with AgNWs, were tested respectively. As shown in Fig. 8(e), it is noted that the sensitivity of the microstructured sensor with AgNWs was 0.831 kPa^{-1} in the low pressure range, which was higher significantly than that of the other two sensors. The sensor has a broad application prospect in the area of e-skin and human-machine interaction. Guo et al. [74] selected carbon nanotubes as fillers for the microstructured dielectric. The sensitivity of the sensor varies with the change of aspect ratios (AR) of CNTs. When the AR of CNTs is 1250-3750, the sensitivity of the sensor was measured up to 2.90 kPa^{-1} in the pressure range of 0-450 Pa and 1.87 kPa^{-1} in the pressure range of 450-850 Pa, as shown in Fig. 8(f), and it's higher than that of the sensor based on pure PDMS dielectric layer. The experiment verified that the sensor was able to monitor the pressure distribution and Morse code.

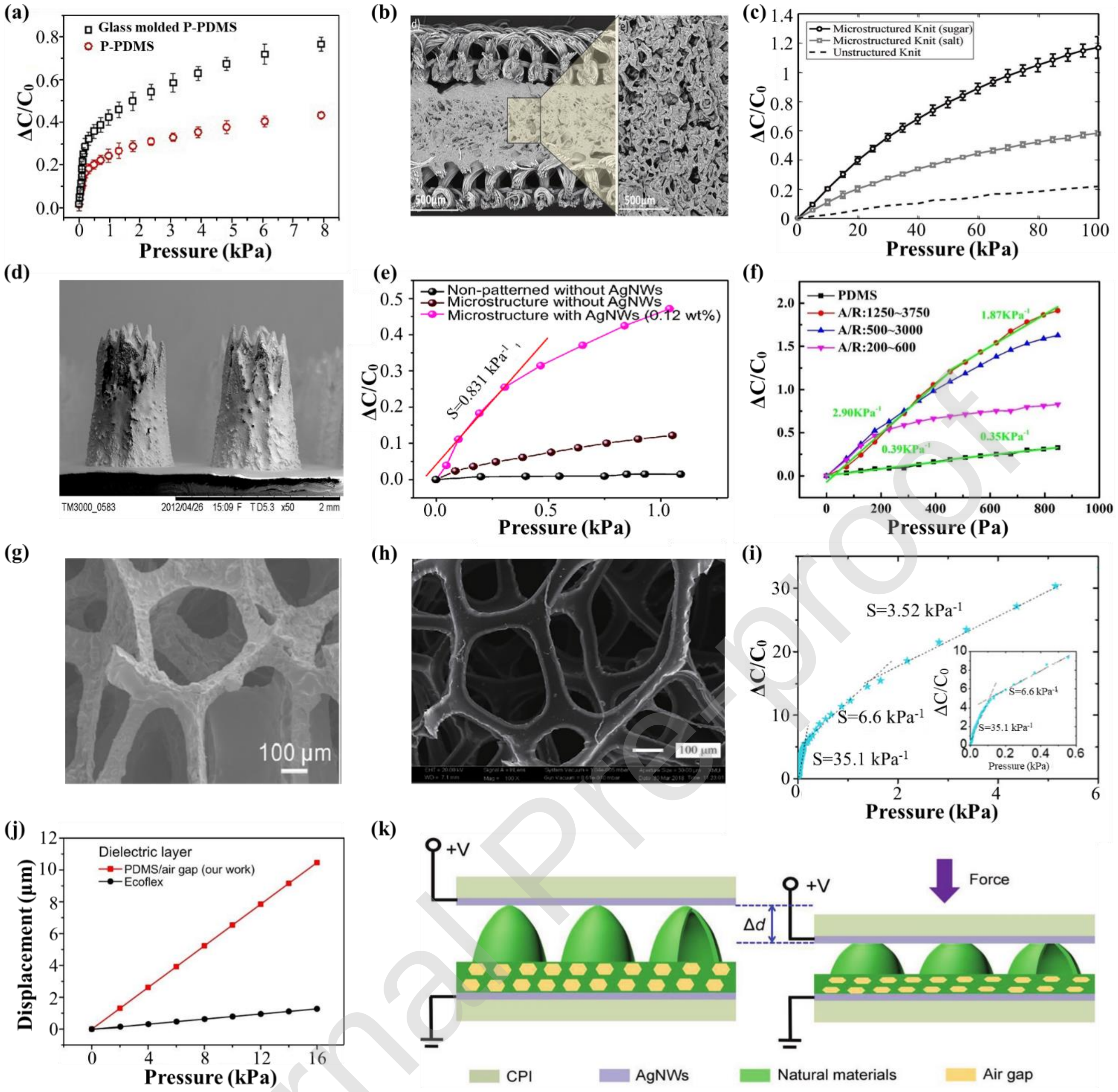


Fig. 8. Methods to improve the sensitivity with measured results, (a) capacitance change ratio of a sensor with a microstructured dielectric and a sensor with unpatterned dielectric, (b) cross-sectional SEM images and partial enlarged views of the dielectric, (c) relative capacitance change-pressure curves of sensors with different materials, (d) side views of a double-stage microstructure, (e) sensitivity of sensors with different dielectric layers, (f) capacitance change of sensors with composite dielectric layer based on various aspect ratios of CNTs under pressure, (g) polyurethane sponge coated with GNPs/MWCNTs/silicone rubber, (h) microstructure of the polyurethane sponge containing ionic liquid, (i) sensitivity of sensor in different pressure range, (j) displacement change of sensors with different dielectric layers under pressure, (k) schematic diagram of sensor before and after compression.

Zheng et al. [101] proposed a new way to improve the sensitivity of sensor by adding materials with high dielectric constant into the porous dielectric layer. In the study, a sensor was prepared using a composite dielectric layer with porous MWCNTs and Ecoflex. Under the synergy of micro-pores and MWCNTs, the dielectric constant of the composite dielectric was significantly improved. As a result, the sensor with MWCNTs/Ecoflex composite dielectric layer has a sensitivity of 2.309 kPa^{-1}, which is larger than the previous related studies constructed with various porous elastomers [28, 66, 94, 102]. It was proved that the sensor can detect pulse and heartbeat of human body, and it can be used in medical measurement device and human-machine interface system. Another kind of sensor [103] was fabricated by impregnating 3D porous polyurethane sponge (PS) into the mixed solution of 2D GNPs/ MWCNTs/silicone rubber to obtain the GNPs/MWCNTs/ silicone rubber/PS dielectric layer with 3D porous structure. As shown in Fig. 8(g), the pore surface of PS is completely covered by the synergistic conductive network. The sensor has the characteristics of high dielectric constant, good elasticity and high sensitivity, and can be used to monitor many physiological activities such as pulse, respiration, muscle movement and so on.

Later, Yang et al. [104] distributed a kind of ionic liquid onto the backbone of polyurethane sponge, as illustrated in Fig. 8(h). The interface capacitance between the ions from the liquid and electrodes confers the sensor with a greater capacitance change and a higher sensitivity under pressure than most existing capacitive sensors, for instance, the liquid metal based senor with microfluidic dielectric layer [105] and the silicon based sensor with air dielectric layer [106]. Then, Pruvost et al. [107] added carbon black (CB) solution when preparing a porous PDMS to fabricate the CB particles evenly distributed on the pore surface of PDMS, and the porous CB/PDMS composite dielectric was obtained. The sensor using the composite dielectric shows ultra-high sensitivity without using transistor to amplify signals. As shown in Fig. 8(i), the sensitivity was measured to be 35.1 kPa^{-1} at 0-0.2 kPa, and 6.6 kPa^{-1} at 0.2-1.5 kPa, which is much higher than the values in other studies, such as the sensor with no amplification and a microstructured PDMS film (0.55 kPa^{-1}) fabricated in Ref. [17] and the sensor with an organic field-effect transistor (OFET) configuration and a pyramid microstructuration (8.2 kPa^{-1}) prepared in Ref. [108]. Owing to the recognizable signals, the sensor has been applied in the measurement of diastolic and systolic peaks of an arterial pulse wave.

In addition to the modification of structure, the other properties of the sensor components can also be explored. For example, the compressibility of the dielectric layer plays a key role in improving the sensitivity. Based on this point of view, Pyo et al. [42] chose air as the main dielectric medium. The air gap between the electrodes of this sensor is larger than that of the sensor with microstructured dielectric layer. The bigger air gap can not only improve the compressibility of dielectric, but also reduce the effect of viscoelasticity on sensitivity like the commonly used elastomers. Here, the sensor was compared with another sensor fabricated using Ecoflex dielectric layer with very low elastic modulus. As displayed in Fig. 8(j), more deformation is facilitated owing to the air gap when the sensor is under loading, inducing a dramatic increase in capacitance. Besides, the low dielectric constant of air results in a small C_0 value of the sensor, which also leads to a larger increase in capacitance. The sensitivity of the sensor was measured to be 0.0655 kPa^{-1}. In some previous studies, the plant surface was used to prepare the template of the dielectric microstructure. However, Wan et al. [109] developed a new capacitive flexible pressure sensor by directly using rose petals as the dielectric. The sensor is composed of the AgNWs/CPI electrode and the dry rose petal dielectric layer, as shown in Fig. 8(k). The dielectric mainly consists of cell walls, the cavity of cell walls and the air gap, giving the sensor a sensitivity of 1.54 kPa^{-1} that can be used in e-skin area.

5. Conclusions and perspectives

This paper reviews the recent advances of capacitive flexible pressure sensors, especially the ways of how to improve the sensor sensitivity. Several commonly used methods have been discussed and summarized from the view point of materials and fabrication of structures. The methods of enhancing sensitivity of sensors were carried out in terms of microstructure, porous materials and composite dielectrics. Among them, to construct microstructure and porous materials mainly gives rise to an increase of the capacitance change by reducing the Young's modulus of dielectric layer and increasing the dielectric constant under pressure, while to fabricate a composite dielectric mainly employs a way to improve the dielectric constant of the dielectric layer. These studies reported that the sensitivity of pressure sensors have been improved effectively, resulting in high potential use of the pressure sensors in e-skin, medical monitoring, human-machine interaction, etc.

Nonetheless, the above methods have their own shortcomings: a) Introduction of microstructure in sensors only improves the sensitivity at a low pressure range, and the sensitivity decreases rapidly when the microstructure is completely compressed under high pressure. b) The relative dielectric constant is improved by adding fillers into the dielectric layer. The sensitivity has been limited improved at a low level. 3) The porous structure, like the microstructure, is difficult to ensure sensitivity under high pressure.

With the increasing requirements of some sensing devices, the development direction of flexible pressure sensors with high sensitivity are proposed mainly including the following points: a), novel sensing materials should be explored with good compressibility, high dielectric constant and low viscoelasticity. b), a sensor with high sensitivity is necessary to be developed for a large pressure range or a sensor with adjustable pressure range. Although the sensor used in the high-pressure range has less strict requirements for sensitivity, it’s crucial to develop a sensor with high sensitivity in a large pressure range in order to expand the application areas. c), to develop a sensor that can achieve multi-signal monitoring is a major challenge. In addition to the detection of pressure, monitoring of other signals such as temperature and humidity are essential for sensors, for example, sensors in e-skin. To date, although the performance of developed sensors can not meet the requirements of various fields, it is believed that the progress of new materials and technology will give a continuous development of sensors with substantive breakthrough, and bring the sensor with more potential use in areas of e-skin, medical equipment, human-machine interface and so on.

Conflicts of Interest: The authors declare no conflict of interest.

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