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Recent progress in flexible capacitive sensors: Structures and properties

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A B S T R A C T

The future intelligent era that will be brought about by 5G technology can be well predicted. For example, the connection between humans and smart wearable devices will become increasingly more intimate. Flexible wearable pressure sensors have received much attention as a part of this process. Nevertheless, there is a lack of complete and detailed discussion on the recent research status of capacitive pressure sensors composed of polymer composites. Therefore, this article will mainly discuss the key concepts, preparation methods and main performance of flexible wearable capacitive sensors. The concept of a processing "toolbox" is used to review the developmental status of the dielectric layer as revealed in highly cited literature from the past five years. The preparation methods are categorized into types of processing: primary and secondary. Using these categories, the preparation methods and structure of the dielectric layer are discussed. Their influence on the final capacitive sensing behavior is also addressed. Recent developments in the electrode layer are also systematically reviewed. Finally, the results of the above discussion are summarized and future development trends are discussed.

1. Introduction

With artificial intelligence gradually entering thousands of households, the field of flexible electronic wearable devices has also entered a new era [1–3]. The rapid development of visualization technology, as well as the huge market of the smart windows and the Internet of Things industry have put forward higher requirements for flexible pressure sensors [4], requirements such as high performance, adaptability, light weight and mass production, etc. Traditional metal/ceramic sensors have shortcomings of inflexible rigid materials and narrow sensing range [5–7]. Polymer-based composites with good flexibility could pave the way for implantable devices for human bodies and stretchable devices. Flexible pressure sensors can be divided into piezoresistive [8,9], capacitive [10], piezoelectric [11] and triboelectric [12] based on their working principles. Compared with piezoelectric and triboelectric sensors, piezoresistive and capacitive sensors have been extensively studied, and it is easy to see many similarities in their mechanisms [13]. In addition, they could have a greater response to static stimuli, and their working principles and manufacturing processes are relatively simple. In the future, the scope of their application could be further broadened. As far as triboelectric and piezoelectric sensors are concerned, their obvious advantages are that they do not require an external power supply system and are more sensitive to dynamic stimuli [14,15].

In addition, the popularity of 5G smart terminals has also promoted the birth of flexible wearable pressure sensors that can achieve the characteristics of "human-centered and human-computer interaction" [16,17]. A pressure sensor that can feed back the external force stimulus as the output of electronic signal change of various materials has been favored by researchers. Integrating the above-mentioned embedded flexible sensors into a human body can project its instructions and corresponding movements more accurately and intuitively. These pressure sensors have important applications in systems such as healthcare [18], human-machine interface [19], soft robots [20,21], and smart homes. When the sensor performs the monitoring of body movement and sends complex commands, it has also exposed some aspects that need improvement. For instance, while sensing external stimuli, it is also expected to detect instantaneous and continuous operations such as inertia, vibration, normal force, and shear force [22–24], so as to achieve the requirements of multi-functional seamless interaction.

Compared with pressure sensors manufactured on other principles, capacitive pressure sensors have received extensive attention because of their simple structure, wide monitoring range, good dynamic response, and low power consumption [25–29]. In the latest research, many have focused on another advantage of capacitive sensors: the charges accumulated on the charged plates are easily affected by the surface charges of other objects, meaning that sensing could be realized without direct contact [30].

This review will discuss in detail the relevant research progress of

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pressure sensors based on the capacitive mechanism. Fig. 1 shows four different dielectric layer structures and key processing methods of capacitive sensors summarized based on the concept of "toolbox". A summary and forecast will also be given regarding the future development trend of capacitive sensors integrated into artificial intelligence and human-computer interaction.

2. Working mechanism of capacitive pressure sensor

2.1. Working mechanism

Capacitive sensors are defined by the fact that the capacitance changes under external stimulation [31]. A common capacitive sensor is a parallel plate capacitor, which is usually separated by an insulating dielectric layer, sandwiched between upper and lower conductive electrodes. The equation for the capacitance is as follows [32–36]:

$$C = \frac{\varepsilon_r \varepsilon_0 A}{d} \tag{1}$$

It can be seen that the capacitance is affected by three factors: the facing area of the two conductive electrodes (A), the distance d between the two plates (d), and the relative permittivity e of the dielectric layer (ε) [37–39].

Under a mechanical external force, the geometric shape between the two polar plates changes, and this leads to a change of the facing area and the distance, so that the capacitance also changes. This is the variable area and distance type [33,35–37]. Of course, because of the change of distance (d), the network between the dielectrics also changes, resulting in a change in the dielectric constant, such that this is called the variable dielectric constant type [32,34,39].

2.2. Key parameters of capacitive pressure sensitive sensor

The key to successful development of flexible wearable devices is to have excellent sensing properties, such as high sensitivity [28,40], wide response range [41] and good cycle stability [42]. For capacitive pressure sensors, the response range is between the upper and lower limits of its test capability. The polymer matrix used in the dielectric layer, the type and size of the nanoparticles, and the interaction between the two, as well as the preparation method, all determine the stretchability of the device.

Sensitivity is one of the issues of most research interest in wearable devices. The main variable that controls sensitivity in capacitive sensors is the relative change of capacitance under a certain pressure. The sensitivity of a parallel plate capacitor is given by the formula (2) [25,43]:

$$S = \frac{\Delta C / C_0}{\Delta P} \tag{2}$$

S represents the sensitivity, ΔC is the relative change of capacitance, C_0 represents the initial capacitance when no pressure is applied, and ΔP is the change in applied pressure. From the formula point of view, in order to increase sensitivity, it is necessary to increase the relative change of capacitance as much as possible while keeping the applied pressure range unchanged. In order to obtain a higher sensitivity of the sensor during compression, many studies have adopted porous structures [44], including sponges or foams [45], and some microstructures [46,47] to improve the deformation behavior of the system.

A faster response time to external forces is a critical issue. If the signal lags, the sensor would perform poorly under long-term operation [48]. The hysteresis in monitoring physiological signals is mostly caused by the

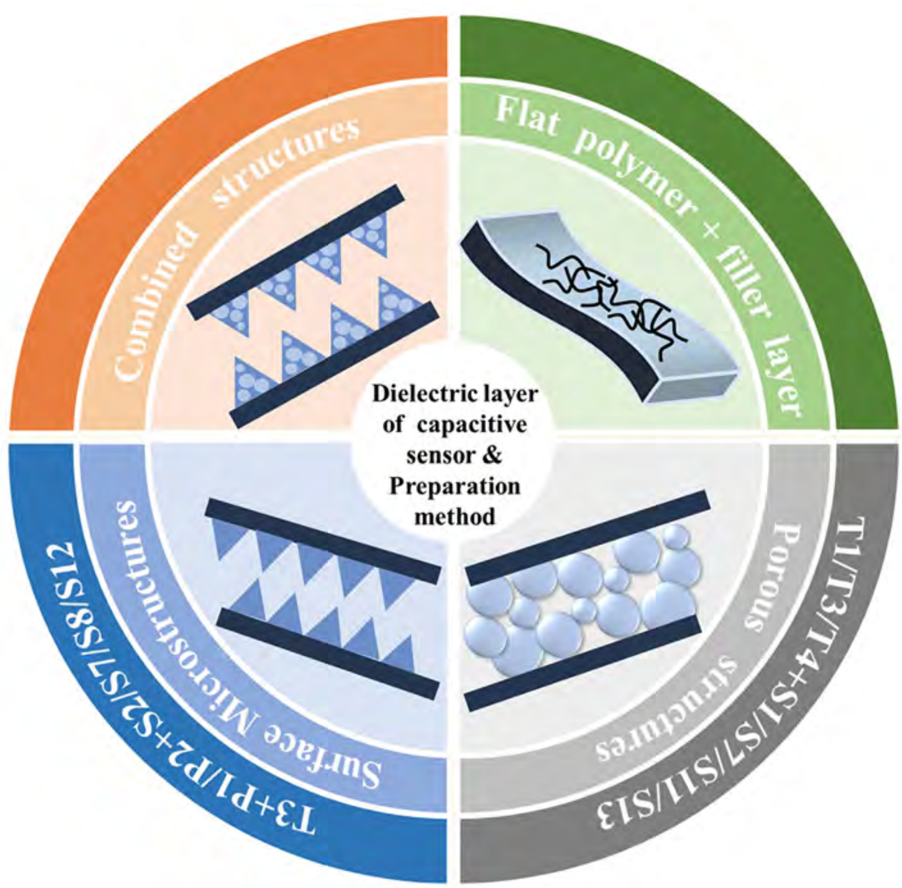

Fig. 1. Schematic showing four different structures of dielectric layers for capacitive sensors and the key methods, based on the Toolbox concept, used to manufacture them.

viscoelastic properties of polymers. Under an external force, the elastic deformation of a polymer cannot keep up with the change in external force, and thus lags behind the strain, so the corresponding time is different [49]. This phenomenon often occurs when the binding force between rigid nanoparticles and the elastic matrix is not very strong, and the nanoparticles cannot completely return to the original position after strain is applied, and two-phase separation occurs, resulting in signal delay. This is especially common in carbon-based composites. Strengthening the interaction between polymer and nanofiller can effectively improve the hysteresis and make the sensor more durable [42,48–50]. Zheng et al. have proposed that good interfacial contact and interface engineering is beneficial for suppressing hysteresis and improving stability. For organic thin-film transistors (OTFTs), they adopted a type of sandwich structured dielectric or polyelectrolyte material for improving the performance parameters of OTFTs at low voltage [40,48,51].

3. Design strategy for dielectric layers

To fabricate an integrated system of flexible and stretchable capacitive sensors for wearable health monitoring usually includes: (1) conductive electrode [52]; (2) flexible/stretchable dielectric layer [43,49]; (3) occasionally including packaging materials [18].This section will introduce the electrode materials and dielectric layers of capacitive sensors in detail. In this process, the concept of " toolbox" is proposed to summarize typical processing methods and characteristics of materials from around 100 highly cited papers in the past five years, as shown in Fig. 2.

3.1. The concept of "Toolbox"

Traditional sensors based on rigid metals and semiconductor materials have inspired wearable devices in terms of materials, structures, and methods, but the issue of flexibility and consistency of attaching to the human body needs to be solved for high-precision monitoring [53–55]. In recent years, studies have adopted various preparation methods to fabricate different dielectric layers. These new methods have an important influence on the sensing behavior of dielectric layers, and can endow wearable sensors with high flexibility, stretchability, high sensitivity and wide response range [56,57]. Nevertheless, the overall relationship between the preparation method, structure and properties has yet to be systematically discussed. Therefore, the concept of "Toolbox" is adopted to summarize the highly cited literature from the past five years. The preparation methods are divided into three categories: "Type of Processing", "Primary processing" and "Secondary processing". In this way the processing method-structure-property relationship can be systematically discussed. Different combinations of processing tools can be discussed regarding their application to prepare a range of dielectric layers. We summarized 100 representative papers from the recent five years for capacitive sensors. The proportions of literature for combination of processing methods; the types of most used tools; the type of processing; the primary processing in 100 papers is shown in Fig. 3. We believe that these specific statistical results are expected to provide researchers with new ideas and strategies for preparing capacitive flexible sensors. For example, the tool combination is mostly T3+S7, as shown in Fig. 3a. This is because on the basis of solution mixing and followed by secondary processing method (coating), the surface microstructure design can be easily achieved to improve sensitivity. The increase in sensitivity can also be obtained through other secondary processing methods including electrostatic spinning, foaming or 3D printing, etc. However, these methods still need more specific attention. The disadvantage of using more than one type of these secondary processing methods during preparation is that it means a more complicated procedure and offers low production efficiency. The applicability of different processing methods under different scenarios is another issue needing careful attention. The type of processing is also mostly limited to T3. It seems that there is still plenty of room for exploration in terms of new preparation methods for dielectric layers. Adjusting the preparation process to melt processing or in-situ polymerization could bring more possibilities for such preparation. It is worth considering.

3.2. Structure of dielectric layer

In order to realize elasticity for the dielectric layer, a flexible substrate is often used. This is mainly from elastic polymer films, rubber, sponge, or foam [44–47,58,59]. Such substrates are widely used in flexible electronics because of their good mechanical flexibility. Generally, in the dielectric layer of a capacitive sensor, an elastomer is often used as the core dielectric material because of its elasticity and compressibility [60]. As mentioned above, the viscoelasticity of elastomers will bring about obvious hysteresis and a rather long relaxation time. To further improve the sensitivity and working range of the sensor, porous structures have also been adopted to reduce viscoelastic behavior to enhance compressibility [61]. In addition, constructing regular or irregular surface microstructures or micropatterns on the surface of elastomers, as well as combining porous structures with microstructures/micropatterns, can also play an important role in adjusting the sensitivity or response range [62,63].

We will now review the micro-engineering design for the dielectric

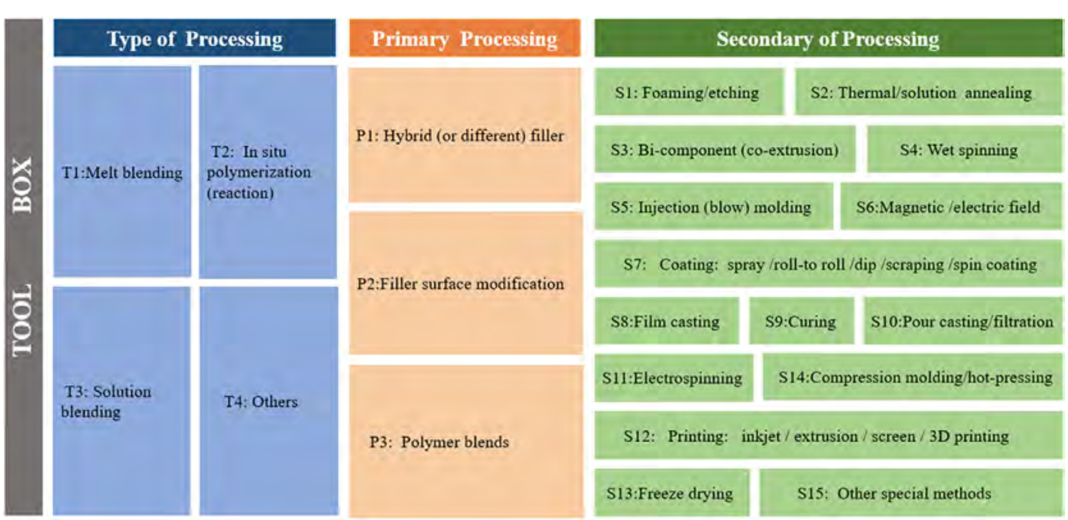

Fig. 2. "Toolbox": the type of processing and specific tools used during processing for dielectric layers of capacitive sensors. These tools are summarized from these 100 paper collected on capacitive sensors.

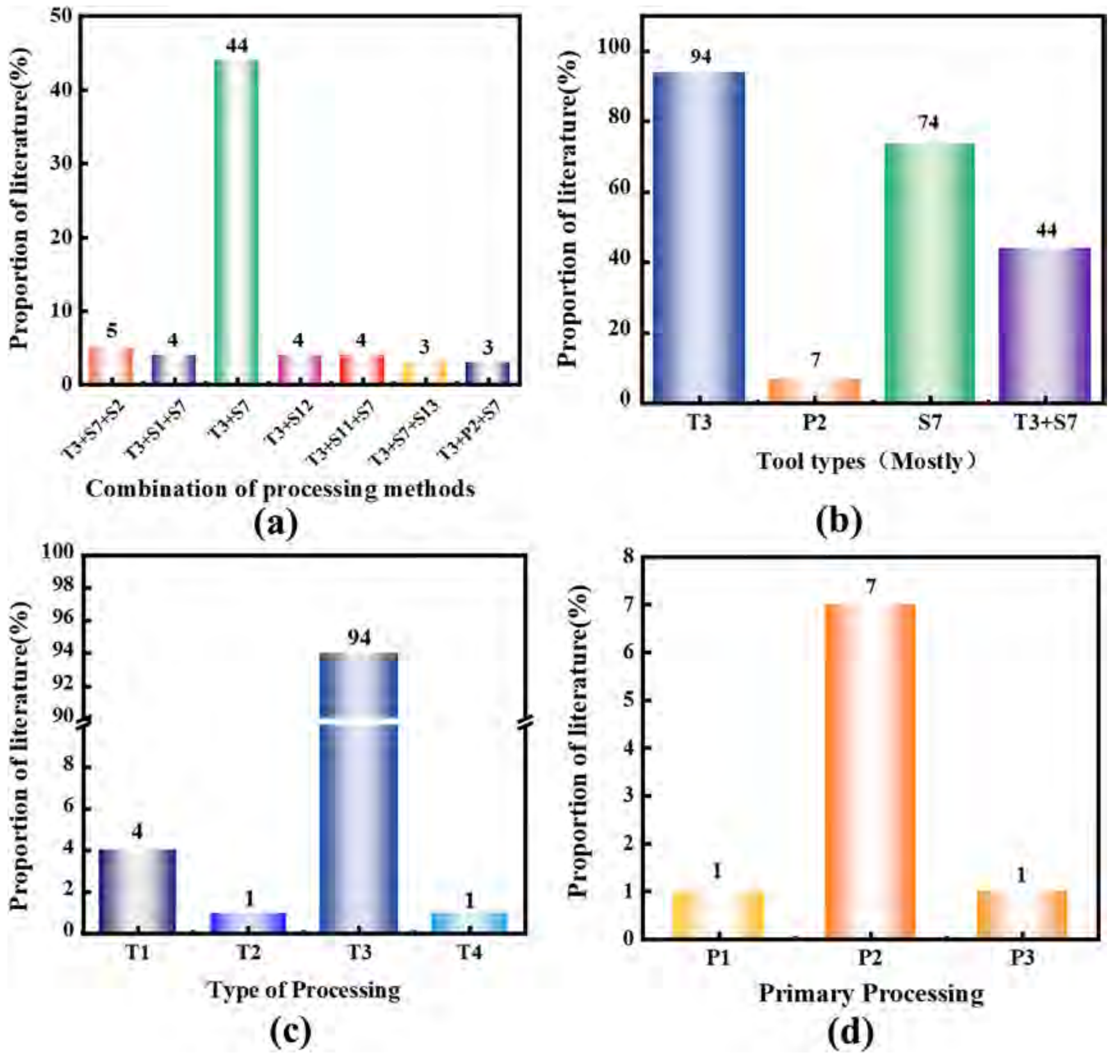

Fig. 3. The proportions of literature for combination of processing methods (a); the types of most used tools (b); the type of processing(c); the primary processing (d) in 100 highly cited papers.

layer and discuss their characteristics in terms of sensing performance. According to the dielectric layer designs reported in the literature, they are divided into 1) flat [63,64]; 2) elastic porous structure [65]; 3) surface microstructure [66]; 4) combined design [67]. We will discuss in detail the scalability, consistency, adjustability, and simplicity issues that these design methods need to face when manufacturing wearable devices. In addition to the performance of traditional sensors that can be referenced, the performance that could be achieved using these design methods and routes in the future will also be discussed.

3.2.1. *Fabrication of a flat dielectric layer*

A flexible pressure sensor relies on flexibility, which can make the electronic skin adhere well to non-planar surfaces and protect the sensor from damage due to large or long-term deformation. At present, using polydimethylsiloxane (PDMS) [68,69] polyurethane elastomer (TPU) [30] a flexible substrate is a popular method for fabricating ductile pressure sensors, as shown in Fig. 4. In addition, epoxy resins with strong adhesion have also attracted much interest [70]. Choosing different crosslinking agents for curing can make it easier to use in different application scenarios. Similarly, in capacitive pressure sensors, electrical insulating polymers such as polyethylene terephthalate (PET) [71], polyvinyl alcohol (PVA) [72,73], and polyvinylidene fluoride (PVDF) [74,75] can also be used as a flexible and stretchable matrix. From the existing literature the most popular among these flexible substrates is PDMS. This is because its preparation process is extremely simple. PDMS has the advantages of good biocompatibility and high extensibility, as high as 1100%. In order to improve its adhesion with attached components, surface modification is often used [76–79]. Common methods include plasma treatment, ultraviolet irradiation, and functionalization of hydrophilic groups onto various surfaces [80,81].

For capacitive wearable electronic devices, if the traditional parallel plate capacitor structure is to be applied over a rather large area, it must be able to uniformly apply the sensing element on the entire surface of the target substrate. In another words, a flat dielectric layer is often preferred. Such a dielectric layer is often prepared by coating. In most earlier flexible capacitive pressure sensors, researchers tended to use neat polymers as the dielectric layer. Earlier work had shown that polymer sheets with a thickness of less than 2 mm is suitable for ultra-thin functional devices [63,64,82,83]. For instance, Han et al. developed an ultra-thin pressure-sensitive and bending-sensitive capacitive sensor. The 1.4 μm-thick PET serves as the dielectric layer, and a patterned AgNWs dispersion is spin-coated as the electrode layer [84]. However, in this type of system, the factor that can cause the capacitance change is very limited. Because the deformation of the dielectric layer is caused by the external force compressing the dielectric layer, its thickness decreases and the capacitance changes. Furthermore, in practical applications, for example, a large pressure range would cause the hardening of a dielectric layer composed of pure polymer, so that the thickness change would not

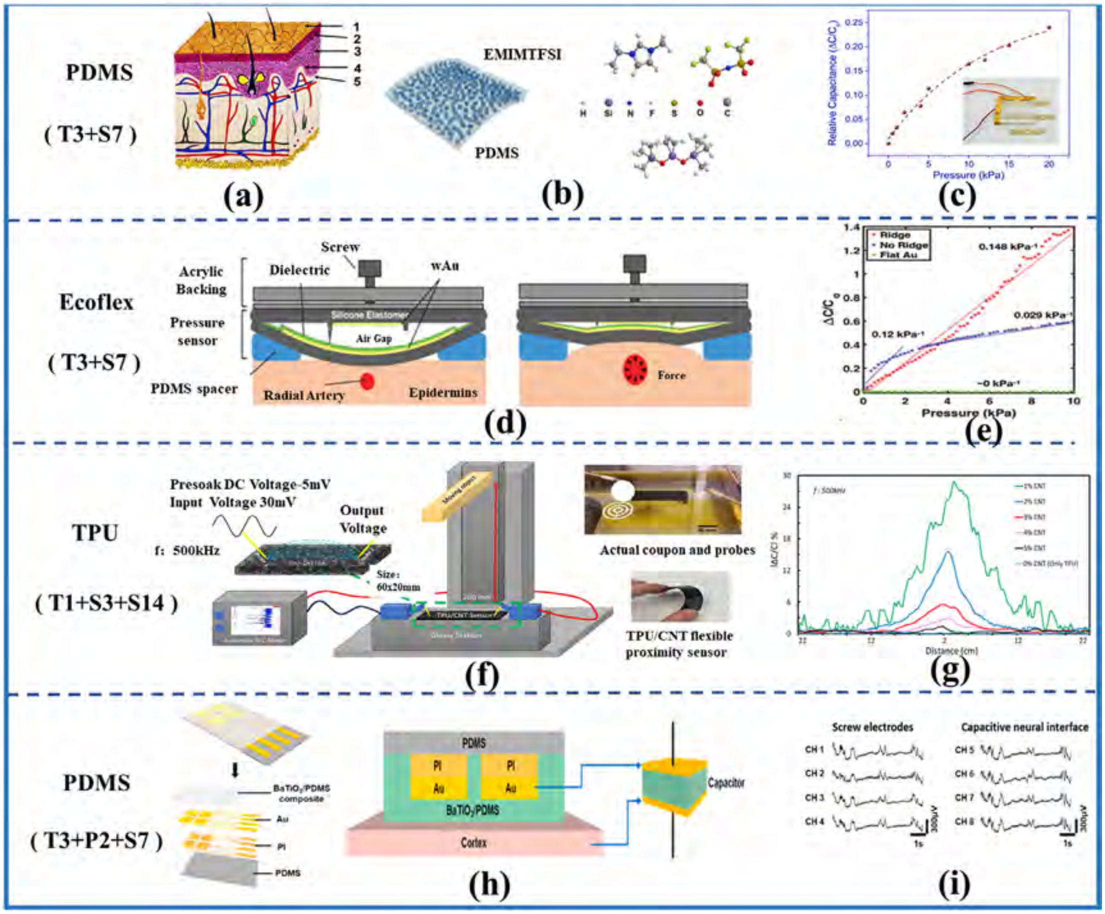

Fig. 4. (a) Schematic diagram of human skin structure. (b) Schematic of separating the spherical phase of the liquid filler in the PDMS matrix. (c) Plot of the sensitivity of liquid-elastomer. (d) Schematic diagram of placing the pressure sensor above the wrist. (e) Plot of the pressure sensitivity curve of the sensor at 0-10kPa. (f) Schematic illustration of the setup of measuring the TPU/CNT proximity sensor using the detection station. (g)Plot of measurement results of the sensitivity i.e. the absolute ratio of capacitance to the initial capacitance, with respect to the distance under different CNTs. wt% are also presented on the right. (h) Schematic diagram of the soft-capacitance neural interface with a dielectric coating made of BT/PDMS composite material and the equivalent capacitance formed by combining it with the cortex. (i) Plot of the ECoG signal recorded by the soft capacitance neural interface (BT:25%).

significantly affect the sensitivity output [85,86]. Of course, this phenomenon may also affect the response of the capacitive sensor from linear to nonlinear, and data accuracy and reliability will be greatly compromised [87,88]. In order to solve this problem, Li et al. suggested doping conductive fillers into the polymer matrix. This is commonly used to improve sensitivity and linearity, because the dielectric constant increases sharply when the filler content is close to the threshold permeability [89]. The design strategy is adding 21.4%, 23.1%, 26%, 27.8% and 23% spike nickel to PDMS to prepare composites as the dielectric layer of the capacitive sensor. Because of the existence of the sharp structure on spike nickel, a linearity of $R^2 = 0.999$ for a wide pressure range up to 1.7 MPa is illustrated. A mixture of 29% spike nickel and PDMS was poured into pre-washed zebra leaf and sandpaper attached to the petri dish. Then, a spike Ni/PDMS composite with a microstructure on the surface is obtained. The composite film not only retains good linearity, but also has a sensitivity as high as 1.149 (while P = 0-20kPa).

3.2.2. Fabrication of elastic porous structured dielectric layer

As mentioned above, the capacitance increases with increasing dielectric constant. Therefore it is easy to think of using a porous elastomer as the dielectric layer, one which can expel air (dielectric constant = 1) during compression, thereby increasing capacitance and effectively improving sensitivity [90,91]. The use of a porous structure in the dielectric layer of a capacitive pressure sensor device has another advantage in that the introduced air gap can increase compressibility and make it easier to deform the structure [92].

The main strategies for manufacturing the porous media layer as shown in Fig. 5 are:

1) Use electrospinning/wet spinning technology to prepare dielectric layer with porosity [93,94];
2) Use commercial foam as the dielectric layer directly [95,96];
3) Use soluble commercial templates such as sugar, salt or polyphenylene microbeads to fabricate pores [97–99].

Recent research has focused on the development of a pore-making process that is easier to manufacture. Advanced and simple electrospinning or wet spinning technology can be used to prepare a dielectric layer with voids [93,94]. Pan et al. developed an ultra-sensitive all-fabric supercapacitive sensor by combining an electrospun nanofiber layer and conductive fabric into a three-layer sensing architecture [100]. Under a pressure of 2.4 Pa, its sensitivity can be as high as 114 nF kPa^{-1}. However, it is not easy to control the pore size when using electrospinning. So using salts or sugars with different pre-determined sizes to fabricate a porous dielectric layer is proposed [99]. Nevertheless, such a method still has extremely limited control over the final structure. Although most studies will choose to adjust the void-to-void spacing according to the diffusion rate and diffusion range of the two substances in the matrix, it is still very restrictive compared to the mold formed by a photolithography process [101,102]. In the porous template method, soluble microbeads

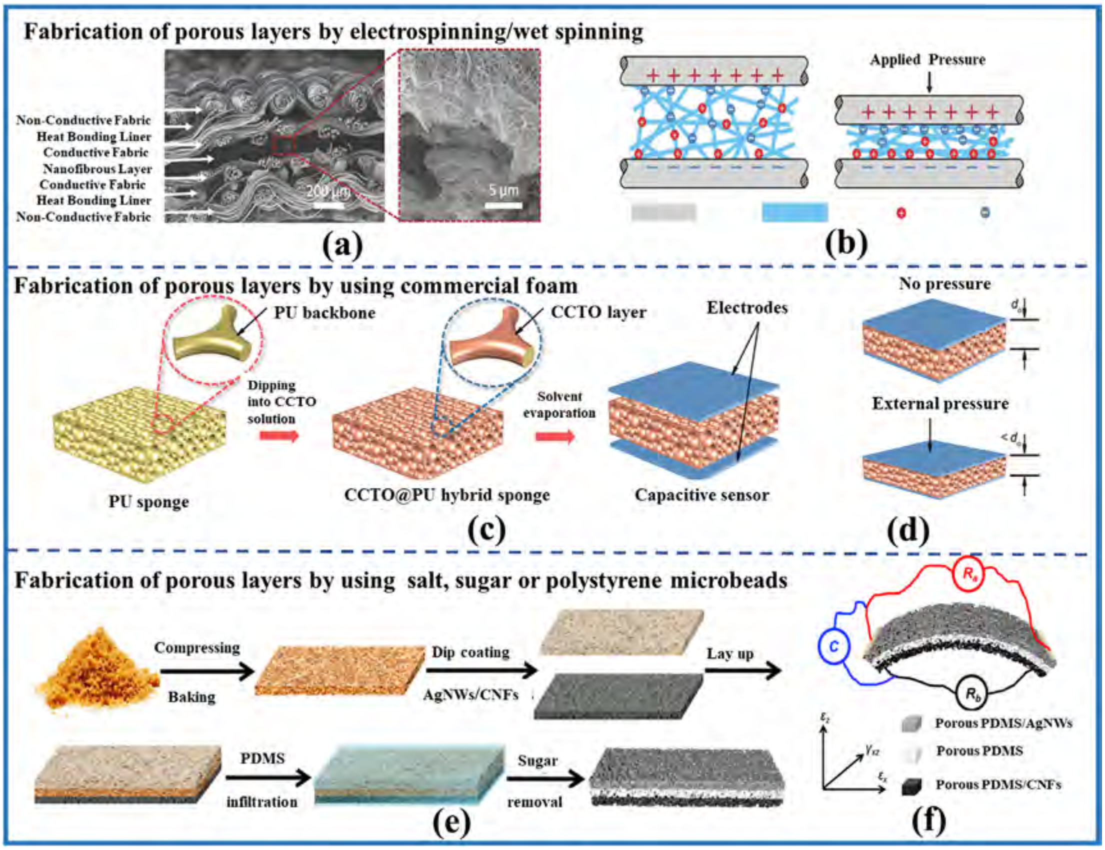

Fig. 5. (a) SEM images of the nanofabric pressure sensor in cross-section views; (b) Operational principle of the nanofabric pressure sensor in which ionic nanofibers are in contact with two conductive fibers. (c) Fabrication of the CCTO@PU foam-based capacitive sensor. (d) Schematic illustration of the sensing mechanism of the sensor. (e) Fabrication of conductive porous layers based on salt templating protocol. (f) A unitized sensor for simultaneous measurement of multiple forces.

are roughly arranged into the shape of the desired array, and the elastomer solution is cast on the array. After the solidification is completed, the initial microbeads can be dissolved easily and neatly to obtain a uniform porous structure.

In addition, the widespread use of commercial porous foams with different porosity can also eliminate the multiple cumbersome steps in the manufacturing process. The commonly used post-processing steps are either dipping the foam into a pre-dispersed conductive filler solution, or coating the foam with an elastomer solution doped with conductive filler [103,104]. However, in the actual application process, it may be necessary to accept differences in internal structure and porosity caused by different batches of foam [97]. This factor must be taken into consideration when realizing the requirements of structural adjustability. At present, there have been many reports on the preparation of flexible capacitive pressure sensors by combining a porous microstructure matrix and functional fillers such as carbon tubes, carbon black and other conductive fillers or dielectric fillers such as barium titanate ($BaTiO_3$) [44,105] and calcium copper titanate (CCTO) [95]. For instance, textiles can provide excellent porous microstructures because of their own texture and pores, and smart textiles can “feel” and respond to the stimuli of an external environment [106,107]. They can be easily integrated into clothing without sacrificing softness and functionality. Chen et al. [108] reported a study based on silver-plated nylon fibers wrapped with polyurethane encapsulated cotton fibers, in which polyurethane-bonded core-spun yarn was used as a strong dielectric layer and silver fibers were used as electrodes. The sensor exhibits excellent capacitance linearity ($R^2 = 0.9975$) and can also have high dielectric stability after 10,000 cycles.

3.2.3. Fabrication of surface microstructured flexible dielectric layer

In capacitive sensors, the dielectric layer is mostly an elastomer with good elasticity and compressibility. The structure and performance of the elastomer can greatly affect the sensing performance of a wearable device. This is because the viscoelasticity of the elastomer would cause significant hysteresis and a longer relaxation time, which are not conducive to sensing performance [25,47]. In order to further improve sensing performance, researchers usually choose to reduce the viscoelasticity of the dielectric layer to reduce relaxation time. Effective measures include the design of surface microstructures [109]. Constructing a surface microstructure can increase the air gap between the two electrodes, making the dielectric layer easier to deform, which can reduce the viscoelastic behavior to better increase compressibility [110]. In addition, surface microstructures with various shapes have been manufactured, including micro-pyramid type [74],micro-dome type [86], and micro-patterning [111]. These surface microstructures exist in a variety of forms such as regular/irregular, symmetrical/asymmetrical, etc. to adapt to different properties.

In Fig. 6, the specific design engineering for the realization of these surface microstructures is shown and includes:

i) Using easily obtainable templates such as sandpaper to produce randomly distributed and uneven surface microstructures. The operation is very simple and inexpensive.
ii) Using nano-sized micro-pattern circuits printed out by systems such as microelectronics to form the required micro-structure plane style.
iii) Using standard photolithography and coating processes, surface microstructures with three-dimensional arrays such as cylinders,

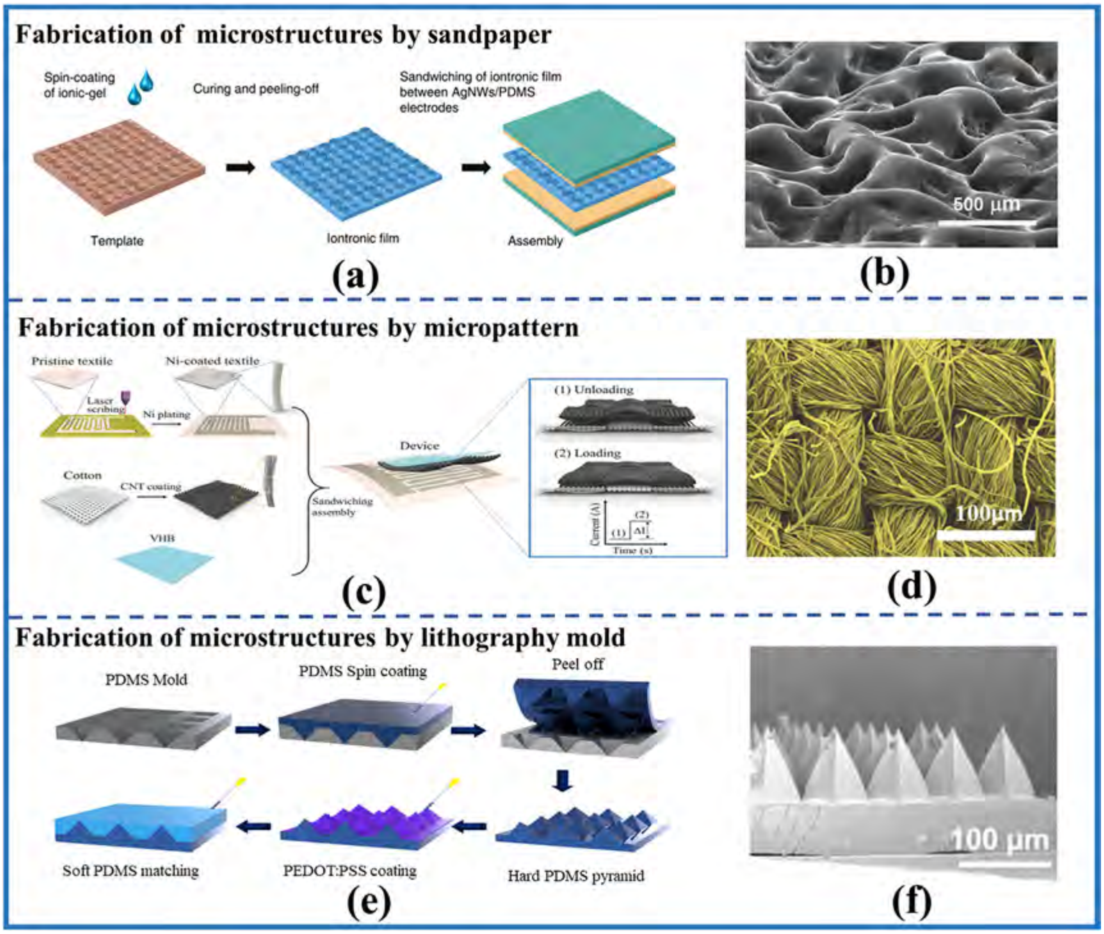

Fig. 6. (a) Schematic of the fabrication process of ultrasensitive interfacial capacitive pressure sensor. (b)The FESEM images of the inclined view of the microstructured film from the sandpaper template. (c) Schematic of the fabrication depiction of textile-based pressure sensors. (d) SEM images of CNT-coated cotton fabric at 100 μm. (e) Schematic of the fabrication sequence of a transparent linear pressure sensor. (f) SEM images of the cross-sectional PDMS pyramid layer.

pyramids, and hemispheres can be etched on silicon or copper substrates.

For sensors with the same substrate thickness, sensor with a surface microstructure can have a smaller initial capacitance than a sensor without one. This is because the dielectric constant of air is 1. Under pressure, the presence of surface microstructure causes the air gap between upper and lower electrodes to be exhausted, which can cause the sensor with the surface microstructured dielectric layer to have a larger capacitance change [112]. Under the same pressure, a smaller initial capacitance and a larger capacitance change is observed. Thus the sensitivity can be higher than other sensors.

As mentioned in Section 3.2.1, a PDMS film is a popular flexible substrate that integrates high-performance nanofillers and flexible sensors [68,110]. Then, building a microstructure on the PDMS film surface can greatly improve a series of sensing properties. The micro-pattern molds are generally cast by spin coating, photolithography, etching, and other processes, and then PDMS is cast or spin-coated on these micro-pattern molds [109,110]. Finally, the surface microstructured PDMS film can be obtained after curing. In addition to the micro-pyramid shape that has been demonstrated by finite element modeling to be able to reduce the effective mechanical modulus of elastomers by about an order of magnitude, bionic biological structures have also been introduced for capacitive pressure sensors [110]. The most popular biological material is the leaves of plants, and the lotus leaf is one of the natural molds with self-cleaning properties and hydrophobicity. Guo et al. [113] reported that these natural mold materials contain micro-powder structural elements, which can be transferred to the film surface after casting the elastomer on the surface and solidifying, and the demolding process is very easy. In addition, Bao et al. [28] developed an ultra-flexible capacitive sensor containing the interesting microwire structure in nature. The interface layer of this micro-hair structure formed by PDMS has been proven to maximize the effective contact between the sensor and the irregular surface of the epidermis. It also can achieve non-invasive conformal attachment to the skin.

3.2.4. Combined structure

Despite the implementation of the above-mentioned design strategies, no elastomer-based capacitive sensor has shown a sensitivity exceeding 10KPa^{-1}. Further studies have shown that when a variety of methods are combined for the preparation of the dielectric layer, such as the introduction of porosity in the dielectric layer with surface microstructure, this helps increase the sensitivity [67]. For instance, Park et al. used PS beads to fill a pyramid structure with a bottom length of 50 μm, and then pre-dispersed PDMS is used to penetrate the gaps between styrene beads for complete curing [47,67]. Finally, they dissolved the styrene beads in an organic solution such as toluene to form a highly layered dielectric layer, and then embedded it on a flexible substrate to complete the sensor. Such a manufacturing method provides a new type of porous pyramidal dielectric layer with a pore size of about 2μm, as shown in Fig. 7. It achieves unprecedented sensing behavior. On the one hand, it can accurately distinguish the number of fruit flies landing on it. On the other, it can achieve a sensitivity of about 44.5kPa^{-1} for a pressure below 100 Pa. Furthermore, the device has been proven to be

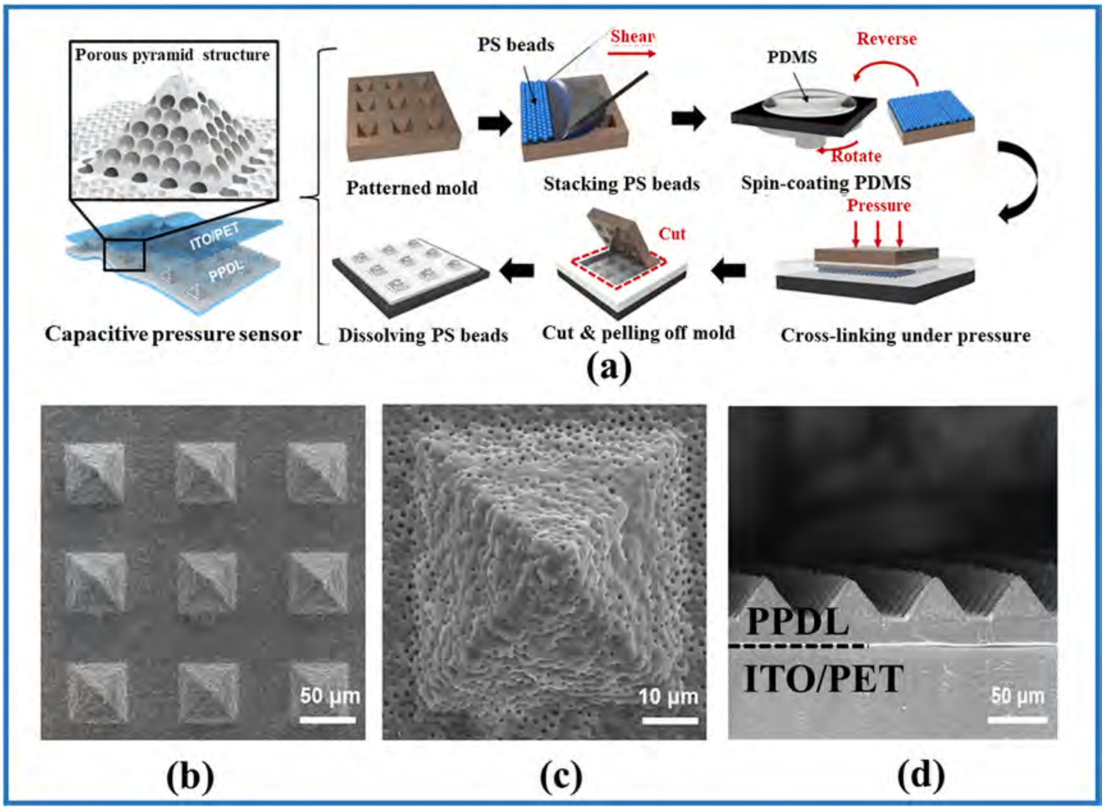

Fig. 7. (a) Schematic of the fabrication depiction of porous pyramid dielectric layer (PPDL). SEM images of the top view of the porous pyramids (b, c) and cross-sectional view of the PPDL(d).

insensitive to temperature changes up to 100 °C and strains up to 60%. This meets the most important feature required of electronic skin, that is that it does not respond to other unintended signal inputs.

Compared with flat films, surface microstructures of different shapes are more likely to be compressed and deformed, but the commonly used method of photolithography is expensive and pollutes the environment. As a result, some plants such as flowers and leaves with specific surface structures have received widespread attention. After in-depth research, Guo et al. [114] found that rose petals, rose leaves and acacia leaves are largely constructed of foam-like hollow structures, and they can be well maintained after supercritical drying treatment. This undoubtedly makes it possible to apply such highly compressible structures to electronic skin. The team used dried rose petals with a uniform papillary microstructure array directly as the dielectric materials and sandwiched them between two flexible electrodes to develop a capacitive electronic skin. The device can work under a load of 0.6Pa-115kPa and exhibits a sensitivity of $1.54kPa^{-1}$. Also, the recognition of human gestures and monitoring airflow can be realized. Such a strategy is also cost-effective and environmentally friendly.

In above examples, it is clear that combining a variety of micro-engineering methods can indeed improve some of the existing micro-engineering deficiencies. For instance, the combined method has more prominent advantages at low pressure. This is because the introduction of the porous element makes the microstructure more easily compressed or even saturated with a small external force. However, the advantages of porous microstructure components over solid microstructures are weakened in the high-pressure range [47,114]. Therefore, when developing capacitive sensors, a design strategy based on actual application goals needs to be carefully considered.

4. Design strategy for electrode layers

When manufacturing capacitive sensors, research mainly focuses on the preparation of the dielectric layer, but the electrode layer as an indispensable part of the sensor is also worthy of detailed study. Generally speaking, conductive materials can be used as electrodes on the sensor [115,116]. However, with the rapid development of smart terminals, higher requirements have been placed on electrode materials. At present, the electrode materials for sensors must also have stretchability to better fit the human skin. Thus the design of stretchable electrodes is now more favored [24]. For example, silver nanowires with high aspect ratio, ultra-high electrical conductivity and excellent transparency have been widely used in various advanced structure electrode materials [24, 115].

Fig. 8 shows the four most popular electrode materials in the 100 highly cited papers: 1) Hydrogel [117]; 2) ITO/PET [118,119]; 3) Copper tape [120]; 4) Conductive network layer [121]. Amongst them, ITO/PET and copper tape have been widely used because they are the most common electrode materials [118,119,122]. Along with the emergence of an endless stream of various capacitive sensors, the types of electrode materials have also increased. Therefore, in the following we will introduce common and newly developed electrode materials with unique characteristics.

4.1. Commonly used electrode layers

In the past ten years, metal (Au, Pt, Ag) and indium tin oxide (ITO) films have been used as electrodes in flexible/wearable devices [119, 122]. Sputtering is the most commonly used process for transferring such metals to a target substrate. Its advantage is that it can directly coat conductive materials such as Au [123], although there is a prerequisite that the target substrate must be flat. Thus the flat dielectric layer discussed in 3.2.1 is more suitable for the sputtering process. There is also a simpler way to fabricate an electrode layer. That occurs where one end of a copper tape or wire is directly connected to the dielectric layer up and down, and the other end is connected to an LCR meter to monitor the

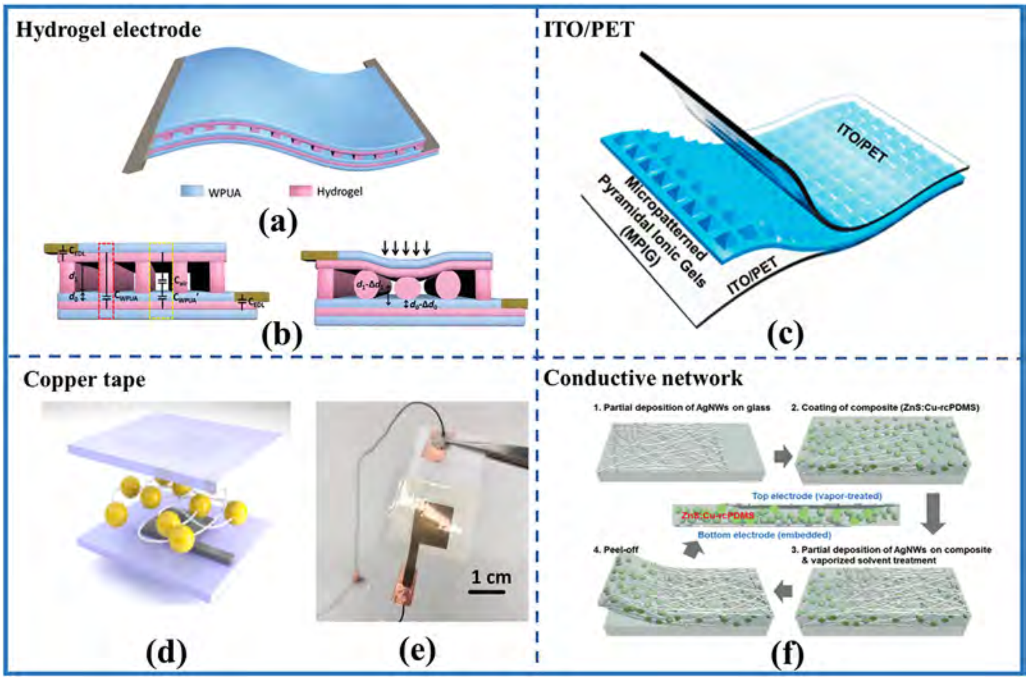

Fig. 8. (a) Schematic of an all-printed capacitive sensor with a 300 μm thick hydrogel electrode layers. (b) Schematic illustration of equivalent circuit of the printed structured device between two hydrogel electrodes. (c) Schematic of a mechanically capacitive pressure sensor with ITO/PET electrode layers. (d) Sketch of the capacitive sensor based on electrospun PVDF nanofibers incorporating insulating microbeads. (e) Digital photo showing a nanofiber/microbead pressure sensor with copper tape. (f) Fabrication procedure of a capacitive photodetector with AgNWs conductive network as electrode layers.

change of capacitance under an external force. The copper tape [124] connected to the instrument can also rely on the excellent conductivity of copper itself to play the role of shielding.

ITO/PET also utilizes a magnetron sputtering strategy. This is carried out by sputtering a layer of transparent ITO conductive film on the PET substrate [118,122]. Because of its good mechanical properties, good folding resistance and inherent transparency, it is often used as a touch screen in mobile communication equipment. When using ITO as the electrode of a wearable electronic device, it is often possible to perform surface modification if needed [125]. For example, most work performs oxygen plasma treatment on the ITO/PET substrate before depositing the elastomer substrate, so that the pre-cured elastic dielectric layer is more firmly attached to the ITO and not easily peeled off during deformation. Another example is a study on a flexible ionic liquid capacitive sensor from Qing et al. [92]. Here the fluorinated separation layer of trimethoxy silane in 3% IPA is used to make the surface of ITO hydrophobic, and thus better reversible contact can be achieved for the ionic liquid and the ITO electrode.

4.2. Specially designed electrode layers

From design point of view, capacitive and resistive sensors have many similarities, including the matrix materials, the type of filler, and the preparation process, although the largest difference between the two comes from the preparation principle [126]. Furthermore, because of the obvious hysteresis of the resistance sensor, the conductive network would change irreversibly under cyclic strain [125,127]. In order to tackle this issue, Pan et al. [100] proposed connecting a variable electric double layer (EDL) nanofabric with ionic fibers in series, and then using the combined structure as a resistance element. It is worth noting that the capacitance of this new supercapacitor sensing structure is about three orders of magnitude higher than that of the traditional parallel plate capacitor, and it is widely used in emerging energy storage and conversion. In such a structure, the resistive element with a conductive network is used as the electrode layer for the capacitive sensor, rather than directly used as the resistive sensor. This method can not only avoid the shortcomings of resistive sensors, but also brings a new strategy for the preparation of capacitive sensors. For a flexible sensor with the classic sandwich structure, the upper and lower layers are replaced by electrode layers composed of a conductive network, and the middle remains a dielectric layer. Such a structure can monitor not only the capacitance signal, but also the resistance signal. Based on this design concept, Wang et al. [97] prepared a sensor that integrates capacitance and piezoresistive mechanisms to quantitatively analyze the mechanical stimulation of the simultaneous applied normal pressure, in-plane stretching, and transverse shear force.

The above-mentioned strategy for the preparation of an electrode layer cannot be further optimized to the overall performance index of the wearable capacitive sensor. Therefore it is particularly critical to integrate various factors, and take both performance and design process into consideration before the experiment [128]. Recently, the strategy of implementing micro-patterning of electrodes has also attracted increasing interest. This design inspiration mainly comes from the micro-patterning of dielectric layers. When the electrode of the sensor is processed using a microstructure, its compressibility can make the upper and lower electrodes closer. Especially compared with the flat electrode without special treatment under the same pressure, the degree of deformation will be more obvious [65]. The processing of the micro-pattern will also increase the contact area of the electrode to further improve the relative change and sensitivity of the capacitance [65,128,129]. This is very similar to the principle of micro-patterned dielectric layers. For instance, Jong-Woong et al. [84] proposed using nano-scale metal structures to be sintered under light irradiation and firmly adhered to a 1.4mm thick polymer substrate to realize the patterned design of AgNWs. Different from the traditional laser ablation method, this new irradiation method can avoid the degradation in physical properties of the polyester substrate. In addition, the interfacial bonding between the AgNWs electrode and the polyester substrate is also shown to be adequate during the application of external force.

5. Outlook and summary

Wearable electronic devices are gaining ever increasing attention. The application for wearable smart terminals has spread from monitoring human body signals to information, medicine and health. As a result, the potential of wearable technology is manifest, and capacitive sensors with executable effect are also attracting increasing attention. Therefore, this entire review revolves around wearable capacitive sensors, which have been discussed in detail from their two core components: the dielectric and electrode layers. The performance of the sensor electronics can be studied through the substrate, filler and structure used for each component. The functional characteristics of the sensor can also be adjusted by appropriate design of the dielectric and electrode layers, and by selecting a suitable manufacturing process. We summarized four types of structures of the dielectric layer, consisting of: doping polymers or fillers into the polymer to design a flat layer, porous structure, surface microstructure, and combination methods. Similarly, for the electrode layer, we also summarized four commonly used materials, namely hydrogel, ITO/PET, copper tape and a conductive network layer. The above design strategies are used to obtain high-performance capacitive sensors. It is also desirable to have multi-functional capacitive sensors, and further development of their multi-functional characteristics can be expected.

Wearable electronic devices are attracting increasing attention. Allowing smart wearable display devices to become part of the body without causing discomfort means that the device must actively incorporate a flexible, foldable and stretchable design shape to overcome the issue of mechanical deformation due to bending and wrinkles. Hence, it is indispensable to use stretchable materials and structures. This type of problem can be solved well by selecting suitable flexible materials and processing schemes summarized above.

Mechanical failure of wearable electronic devices is unavoidable when they are frequently used. Therefore, sensors with self-repairing capabilities have been proposed. The cells of human skin are constantly renewing themselves in a specific cycle, and when the skin is damaged, it can repair itself. So, it is very important to develop a flexible pressure sensor with a self-healing function, just like making the electronic skin a real skin. However, electronic skin is composed of many sensor arrays, so a single sensor must also be high-performance. With the support of high-performance pressure sensors, systems with integrated sensor arrays can more accurately provide comprehensive detection functions within the adjustable sensing range. Therefore, we believe that starting with the preparation of the dielectric layer is the best solution, and the material of each dielectric layer and the corresponding design strategy would have a unique impact on performance parameters. The use of manufacturing technologies such as microstructures can undoubtedly promote the development of high-performance capacitive sensors, but they are also necessary to resolve the contradiction between opposition and coordination in some common microstructure capacitive sensors. The most prominent conflict is high sensitivity and stretchability. When fabricating devices with microstructures, large capacitance changes can be easily obtained at small strains, but when the application scenario is switched to large strains, stretchability is required to maintain the integrity of the structure and for the shape not to be damaged. It is still a challenging task to unify the two. Other issues that need to be considered are cost and the microstructure process. To obtain a uniform and fine microstructure, complex and time-consuming photolithography technology must be used, but the production cost is rather high. Although microstructure based on plant leaves or sugar template can be used, these methods do not control the morphology and uniformity of the microstructure, and this may affect its performance. This may greatly limit the large-scale production and application of sensors with microstructures. We believe that the existence of these conflicts is not insoluble. They are the driving force for the development of multifunctional flexible stretchable capacitive sensors with ingenious microstructures and broader application prospects.

In general, wearable capacitive sensors are only monitoring devices that can sense information from the outside and convert the received information into electrical signal output. When the specific design and the required application are determined, the advantages or limitations brought by the above preparation methods must be taken into consideration and a reasonable choice must be made. Only when the challenge of combining the properties of materials, the structure of devices and ease of preparation is completed, will the real transition from laboratory to practical applications for flexible wearable devices be realized.

Declaration of competing interest

The authors declare no conflict of interest.

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