Flexible pressure sensors with microstructures

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Funding information
National Key Research and Development Program of China, Grant/Award Number: 2020YFC2007101; National Natural Science Foundation of China, Grant/Award Number: 61933002; Natural Science Foundation of Zhejiang Province of China, Grant/Award Numbers: LQ20A020005, LQ19E030003

Abstract
Microstructured flexible pressure sensors featured with good mechanical properties, boosting a variety of sophisticated application scenarios, including electronic skin (e-skin), soft robotics, wearable electronics, etc. This review is very focusing on the recent research progress of microstructured flexible pressure sensors. For better understanding the corresponding devices, different mechanisms, materials, preparation methods are briefly introduced at the beginning. And with highlighting the significance of microstructure for device performance, the microstructures of different configurations (e.g., pyramid, pillar, hemisphere) are introduced and discussed in detail through analyzing the influence of configuration characteristics and material properties. Finally, according to the existing problems in the application, the research directions of flexible pressure sensor are prospected. Currently, catering to the explosive and ineluctable growth of this intelligent world, considerable microstructured flexible pressure sensors have emerged but their development is still at very first stage. In this review, some guidelines and tunable methods are suggested for the microstructured flexible pressure sensors of wide practical use in the future.

KEYWORDS
configurations, flexible, microstructure, performance, pressure sensors

1 | INTRODUCTION

Artificial intelligence (AI) plays a substantial role in education, medicine, finance, and logistics industries. Pressure sensors as a very basic unit, can perceive the external environment, determining the perception accuracy of AI. With the continuous extension of AI applications, such as electronic skin, soft robotics and wearable electronics, etc., pressure sensors adapting to these sophisticated scenarios are highly demanding. While conventional rigid pressure sensors exhibit poor flexibility and stretchability, failing to be conformal with flexuous contour tightly, which affects the measurement accuracy. In addition, such brittle and bulky sensors also cannot be available for high integration and low power consumption. Therefore, pressure sensors are tending to be flexible, miniaturized, and multifunctional.

Flexible pressure sensors can be prepared with microelectro mechanical (MEM) Si islands in which each rigid island is connected with flexible interconnects to form an island-bridge structure capable of stretching and...
bending. This method also can be used to fabricate large area e-skins. However, owning to the intrinsic brittleness of rigid substrate, the stretchability and conformability of devices are still very limited. Thereby, taking advantage of the intrinsic softness, polymers have become the promising candidates for flexible electronics, enabling good mechanical performance of devices conformal to different curved interfaces. In spite of the good conformability and compliance, the polymer-based devices are susceptible to the hysteresis caused by viscoelastic behaviors of materials. Thus, a variety of microstructures based on advanced micro–nano processing technology are introduced to reduce the hysteresis, contributing to the high sensitivity and fast response of devices. To further adapt to some complex applications, multifunctional flexible pressure sensors enabled by the combination of different working principles or the integration with other working units have become another developing trend.

Benefiting from the great efforts made by the researchers across the world, flexible pressure sensors have experienced a great evolution with improved performances including sensitivity, limit of detection (LOD), response speed, etc., based on the enhancement of materials, fabrication methods and different working mechanisms enabling a variety of applications of flexible electronic field (Figure 1). The progress of microstructural flexible pressure sensors in recent years is summarized in this review. The basic working mechanisms of flexible pressure sensors are briefly described in Section 2.
Section 3 classifies the materials used in flexible pressure sensors, and illustrates the function and application of various materials with some representative examples. The micro–nano processing technology used in fabricating microstructured flexible pressure sensors is briefly described in Section 4. In section 5, the structure designs of the flexible pressure sensor are discussed in detail. The typical structures are classified, and the design method of the structures are described from the perspectives of theory, numerical simulation, and comparative experiment. Section 6 describes the applications of microstructured flexible pressure sensor in various scenarios with summarizing the current problems and prospecting the future developing trend in Section 7.

2 MECHANISMS

The pressure sensors based on different mechanisms have corresponding characteristics and are suitable for different working scenarios. The common working mechanisms of flexible pressure sensors are illustrated in Figure 2, corresponding to piezoresistance, capacitance, piezoelectricity, triboelectricity.

Piezoresistive pressure sensors detect the pressure by the variation in resistance of devices during deformation, as shown in Figure 2a. In this type of devices, the composite films comprised of elastic matrix mixed with conductive fillers (i.e., metal nanowires [97] carbon nanotubes [CNTs], [98] and graphene [99,100]) or the intrinsic conducting polymers [101,102] are usually adopted as the functional layer either laid on interdigitated electrode or sandwiched between two electrodes. [103] When the normal force is applied, [101] the contact resistance \( R_c \) changes due to the change in contact area [104] or contact point [105] in the materials. [106] For composite films, the sensitivity \( \Delta R/R \) can be further developed by expanding the tunneling effect [75,107] in which the concentration of conductive fillers is kept near the percolation threshold. Taking advantages of their simple fabrication, good sensitivity and high spatial resolution etc., piezoresistive sensors have been widely used. [71] But due to the thermal expansion of conductive polymer and temperature sensitivity of resistivity for metals, (e.g., the temperature coefficient of resistance at 293.15K for silver \( \alpha_{\text{silver}} = 0.003819/\text{T} \) and for copper \( \alpha_{\text{copper}} = 0.004041/\text{T} \), the resistive of piezoresistive pressure sensors perform high drift over temperature changes [108,109] which limits their measurement in variable temperature environment.

Capacitive sensors are composed of two adjacent conductors sandwiched with a dielectric layer, capable of transmitting the pressure into the capacitance change. The capacitance (\( C \)) of a parallel plate capacitor can be expressed as \( C = \varepsilon A/d \), where \( \varepsilon \) is the dielectric constant, \( A \) is the overlap area of the plate, and \( d \) is the distance between electrodes (Figure 2b). [110] When the normal force is applied on the sensor, the change of electrode distance \( d \) results in a change in capacitance. [111] Similarly, the shear force can also lead to a change in capacitance by changing the electrode area. [112] The sensitivity of the capacitor can be tunable through the material and structure design of the dielectric layer. [113] In addition, the capacitive sensor is equivalent to open circuit with direct current power supply, thus it has lower energy consumption than resistive sensor. Since the dielectric constant of dielectric layer changes with temperature (e.g., at 293.15K, the dielectric constant decreases at a rate of 0.00175/K for polydimethylsiloxane [PDMS] [114] and 0.00025/K for polytetrafluoroethylene [PTFE], [115] respectively), the accuracy of capacitive sensors are affected by temperature. Moreover, capacitive sensors easily suffer from electromagnetic interference and parasitic coupling to the surroundings [116–118] and also perform poor linearity under dynamic state. [12]
Piezoelectric sensors convert force and vibrations into electrical signals via piezoelectric properties of piezoelectric materials. These materials (i.e., polyvinylidene fluoride [PVDF], zinc oxide (ZnO), barium titanate (BaTiO3)) can generate opposite charges on the upper and lower surfaces when the external force is applied (Figure 2c). After removal of the force, the charges return to the initial state without charges. Benefiting from the fast response, piezoelectric sensors are especially suitable for dynamic pressure measurement compared with piezoresistive and capacitive sensors, while the devices are not very suitable for static pressure measurement due to the energy loss caused by the internal resistance of piezoelectric material.

Triboelectric sensors exploit the combination of triboelectric effect and electrostatic effect to detect the change of pressure. As shown in Figure 2d, two rough surface layers with different electronegativity are attached to two parallel electrodes; when the pressure is applied, the two layers contact with each other, and the electric charges transfer from one layer to the other due to the friction; after removal of the applied force, the charges on both sides cannot be completely neutralized due to the gap between two layers, such that the inductive charges accumulate in the electrodes, generating an instantaneous current in the external circuit, which can be measured to reflect the magnitude of pressure. The triboelectric effect has a rapid response and can be widely used in quasistatic and dynamic pressure measurement. However, the electrical performance of the triboelectric sensor highly depends on the electronegativity of materials, and the roughness of the material interfaces can also influence the triboelectric properties.

Other than these four major mechanisms, pressure sensors based on photoelectric or piezophotonic effect are also used in various fields. For instance, the piezophotonicity based pressure mapping can be achieved by optical signals converted from mechanical stimuli through tuning/controlling the electro-optical process. A rapid-response flexible pressure sensor matrix obtained by exploiting the piezophotonic property of ZnS:Mn particles can directly converting applied force into an optical signal. And the transistor structure also as another effective method can be used to increase the current amplitude with high resolution. However, the structure of the transistor is relatively complex, and the stability of the transistor made of polymer material is unsteady.

3 | MATERIALS

Flexible pressure sensors are usually comprise functional layer, electrode layer and substrate layer. According to different mechanisms, the functional layers can be classified into resistive layer, dielectric layer, piezoelectric layer, etc., which play a decisive role in electrical signal. The electrode layer is usually arranged on both sides or one side of the functional layer, responsible for signal output. The substrate layer is mainly used for encapsulation and protection of sensors. The flexibility and low stiffness of the substrate endow the pressure sensor with high response speed to the compression deformation. Although encapsulation by soft substrate can reduce the wave velocity of the applied force, the delay caused by encapsulation (tens of microseconds) is negligible to the total response time (hundreds of microseconds to hundreds of milliseconds). On the other hand, due to the low stiffness of the substrate, the relatively stiff functional layers are isolated from strains under stretching conditions, thereby improving the measurement stability of the device. As seen from Figure 3, elastomeric polymers can be used in substrates, electrodes and functional layers, such as PDMS, polyurethane (PU), polyimide (PI), and etc.; conductive materials can be classified into metal-based materials, carbon-based materials, and conductive polymers, usually used to fabricate electrodes and functional layers; and piezoelectric/piezoelectret materials are used widely in functional layers of piezoelectricity-based or triboelectricity-based pressure sensors.

3.1 | Elastomeric polymers

Owning to the highly tunable elastic modulus, the modulus of elastomeric polymers almost covers the whole biological range from cell to bone, contributing to the compliance and conformability of flexible pressure sensors. Elastomeric polymers of insultation can be used as substrates and encapsulations of sensors or dielectric layers for capacitance-based pressure sensors. And filled with conductive materials, these insulative elastomeric polymers can be transformed to be conductive, which can be adopted as electrodes of sensors or resistive layers for piezoresistance-based pressure sensors. Some typical materials are presented as following.

PDMS performs low Young’s modulus (~1 MPa) and the modulus also can be changed by adjusting the content of curing agent. The low surface energy enables PDMS conformal to different interfaces through the van der Waals force and the plasma can to some degree increase the adhesion force. PDMS also exhibits good biocompatibility with biological tissues and living cells due to its good chemical and thermal stability and nontoxic properties. PI can be very stable under high temperature ranging from 100 to 300 °C, resistant to different chemical solvents.
and robust enough against high mechanical forces. Polyethylene terephthalate (PET) has a high melting point and good mechanical strength owing to the presence of aromatic ring in the polymeric structure, which can also be very stable under high temperature and moisture. Poly(glycerol sebacate) (PGS) is a biodegradable elastomer with Young’s modulus ranging from 0.05 to 2 MPa, suggesting great potential in bioengineering, for example, drug delivery, and in vivo sensing architecture. With the assist of PGS, a biodegradable pressure sensor has been fabricated for pulse wave velocity measurement.

3.2 Conductive materials

Conductive materials are usually adopted for fabricating electrodes in flexible pressure sensors, or combined with polymers to form functional layer, which involving metal-based, carbon-based, and conductive polymers.

Metal-based materials have relatively higher conductivity than that of others. Among metals, Ag performs the highest electrical conductivity (1.6 × 10−6 Ω cm) and good ductility. Owing to the high aspect ratio, silver nanowires (Ag NWs) shows very low percolation threshold. Taking advantages of cost-efficient and large-scale production, Ag NWs have been widely used in stretchable and flexible devices. Ag NW/polymer (i.e., PDMS) composite or mixture can be fabricated as stretchable conductors, exhibiting good conductivity and stretchability. For instance, a highly conductive and stretchable conductor with Ag NWs embedded below the surface of PDMS exhibited stable conductivity of 5285 S cm−1 over a large range of tensile strain (0–50%). With the assist of Ag NWs in active layer, the sensitivity of the piezoresistance-based flexible pressure sensor could be improved to a maximum of 5.54 kPa−1.

Gold nanowires (Au NWs) also have attracted considerable interest as conductive components of flexible devices, owning to their unique chemical inertness and high electrical conductivity. Additionally, they exhibit good biocompatibility as well as they can easily penetrate in cells or tissues with minimum biological damage. For instance, a gold nanowires-based pressure sensor was fabricated by Gong et al., achieving high sensitivity of 41.14 kPa−1 for health monitoring.

Carbon is an important element in nature with sp, sp2, sp3, and other hybrid orbitals, and there are many kinds of carbon-based materials, including carbon black (CB), graphene, and CNTs etc. Carbon-based materials are usually adopted as in active layers or electrodes with flexible polymers or rubbers, attributing to the good elasticity and conductivity of the composites. For instance, CNTs perform high intrinsic carrier mobility (10 000 cm2 V−1 s−1), high electrical conductivity (104 S cm−1), and great mechanical performance (elastic modulus of 1 TPa). The multifunctional sensor with graphene also exhibited high transparency due to the remarkable carrier mobility (∼ 20 000 cm2 V−1 s−1).

Good electrical properties and elasticity can be obtained by doping these inorganic conductive materials into elastomers. However, the performance of conductive polymer composites highly relies on the concentration of conductive fillers, as high concentration leads to high conductivity but sacrificed stretchability. Moreover, the aggregation of fillers, and the matching problems of interfaces between fillers and polymers may also result in poor conductivity or mechanical properties.

There are a variety of conductive polymers that can be endowed electrical conductivity through initiating in situ polymerization of conjugated precursors, including polyacetylene (PA), polyaniline (PANI), poly(3,4-ethylene dioxythiophene):polystyrene sulfonate
Piezoelectric/piezoelectret materials

The surface of piezoelectric materials can be charged when the pressure or tension is applied in a specific direction due to the piezoelectric effect, and the density of the electric charge is proportional to the magnitude of the applied external force. Owing to the piezoelectric effect bridging the gap between electric charge and force, piezoelectric materials are widely applied to piezoelectric flexible pressure sensors.

The ferroelectrics can maintain a certain residual polarization strength after removing the polarization electric field. Therefore, lead zirconate titanate (PZT), ZnO, aluminum nitride (AlN), etc. have good piezoelectric properties. Although inorganic piezoelectric materials have relative high stiffness and brittleness, with thinning of thickness, they can also have good bending performance. In addition, due to the temperature insensitivity of inorganic piezoelectric materials, the measurement accuracy is high in complex environment. For example, a PZT-based sensor exhibited great advantages in sensitivity, frequency bandwidth, and response time, applied to measure human pulses in different areas, such as carotid, radial artery, and the apical region.

The piezoelectric properties of PVDF are attributed to inherent dipole moment of the repeating monomer (-CH2-CF2-), with electrical poling to achieve maximum polarization in the direction orthogonal to the film plane. Due to the mechanical compliance than inorganic piezoelectric materials, PVDF has become the most widely used piezoelectric material in the field of flexible pressure sensor. However, as β phase is the unstable phase, the piezoelectric properties of PVDF will decrease with the increase of time or temperature. Poly(vinylidene fluoride-co-trifluoroethylene) (P(VDF-TrFE)) is a derivative of PVDF, and its piezoelectricity is unsensitivity to temperature. The flexible piezoelectric sensor that consisted of sheets of electrospun fibers of the polymer (P(VDF-TrFE)) have ultra-high sensitivity in the low-pressure regime (0.1 Pa) and fast response to both compressive and bending forces. In addition, oddnylons, vinylidene cyanide copolymers, and polyurea etc. organic piezoelectric materials even boost wider applications compared with PVDF family. Due to their good acoustic impedance matching with water and biological materials (especially human skin), no additional impedance matching layer is required to improve their coupling efficiency, making it easy to develop efficient ultrasonic transducers for underwater and human related signal detection.

Piezoelectrets that are porous piezoelectric polymeric materials with space charges, have been widely studied because of its higher piezoelectric constant than PVDF and better mechanical compliance than piezoelectric ceramic, including polypropylene (PP), fluoroethylenepropylene (FEP), PTFE, cyclic olefin copolymer (COC). Similar to PVDF, these materials have no intrinsic dipole or domain structure, so they are nonpolar as well. The piezoelectric effect of piezoelectrets can be induced by the nonuniform deformation of the space charge layer in the porous films upon compression or expansion. Due to their outstanding space charge storage capacity enabled by porous microstructures, the piezoelectrets show good and stable piezoelectric property. For instance, the piezoelectric coefficient d33 of PP honeycomb film could reach 200 pC/N, which was about one order of magnitude higher than the corresponding value of PVDF (33 pC/N). If the closed holes in PP honeycomb membrane were filled with N2 (100–450 kPa) or N2O (100–140 kPa), the piezoelectric coefficient could be further enhanced to 790 pC/N, which was about 50 times of PVDF. Similarly, the d33 of multilayer piezoelectric film with PTFE porous film as sandwich layer could achieve 600 pC/N. Due to its good piezoelectric properties, piezoelectret-based flexible electronics have been widely used in wearable energy harvesters, pressure monitoring, and health care.

4 | MANUFACTURE

Based on the progress of micro-nano processing technology, electronic devices are successfully tending to become miniaturized, highly-integrated and high-efficiency. The commonly used micro-nano processing technology (e.g., 3D printing, laser writing, and vapor deposition), enables thin, light-weight, and high-sensitivity flexible pressure sensors. Moreover, three-dimensional microstructures introduced into sensors
have been demonstrated to further improve the sensitivity and some representative techniques are presented as following.

Molding as an easy-feasibility technique prevails in fabricating pyramid,[96] hemisphere[75] and pillar[231] structure, exhibiting high efficiency of preparation and good control of construction. The specific preparation process is shown in Figure 4A.[75] The solution of the flexible material (e.g., PDMS) is cast on the template made of rigid material (e.g., silicon) with prepared grooves on its surface. The flexible material can be solidified through temperature,[85] light[271] and other conditions and finally the microstructure can be obtained by peeling the flexible material from the template. The surface of natural structure, such as sandpaper,[265] leaf,[275] and petal,[272] can also be used as templates to mold microstructures with irregular geometries. As for the microstructured devices of small size, the microstructures are vulnerable to the damage of directly peeling from templates by hand. Transfer printing as a high-fidelity method can transfer devices from donor substrate to target substrate with little damage, enabling high-efficiency peeling (Figure 4B).[214]

Microhair can improve the sensitivity and LOD of sensors. As shown in Figure 4C, the gold nanowires (Au NWs) are grown on the PDMS pyramid structure, enabling a high-sensitivity pressure sensor.[215] The surface of PDMS pyramid are modified to be hydrophilic and 3-aminopropyltriethoxysilane (APTES) solution is used for amino modification, rendering the gold nanoparticles (Au NPs) fixed on amino. The spontaneous growth of Au NWs along the vertical direction can be finally achieved immersed in some corresponding solutions.
Different from pyramid, hemisphere, and pillar structures, porous structure cannot be directly obtained with templates. The chemical reaction and physical dissolution are the common methods to fabricate porous structure with tunable pores through controlling the size of sacrificed particles. During the chemical reaction, the heat exotherm and prepared pores are uniform, resulting in higher compressibility and flexibility of sensors. As shown in Figure 4D, the mixture of PVDF solution and ZnO nano particles (NPs) are cast to form PVDF/ZnO composite film, and the porous PVDF film can be obtained with removing the ZnO NPs through the reaction with hydrochloric acid solution. In the physical dissolution process, salt or sugar is often adopted as sacrificed particles mixed with PDMS and other flexible materials to obtain a well-uniform porous structure. The porous structure also can be formed by material decomposition and volatilization.\(^{[85]}\)

Hollow-based pyramid, hemisphere, and pillar structure also can be achieved but with more complicated preparation technology. A spherical hollow structure is presented in Figure 4E.\(^{[208]}\) The liquid ethanol is gasified at high temperature to form a high-pressure area, making the film bent along the concave surface of the template. The hollowed film is obtained after peeling the covering film and template. Compared with the direct pressing with convex template, this pressure-assisted method with the concave template can avoid the film damage caused by uneven pressure.

### 5 | MICROSTRUCTURE

The performance of the flexible pressure sensor have be significantly improved by advanced materials. However, due to the intrinsic viscoelasticity of polymer materials, flexible pressure sensors are still limited by low-pressure insensitivity, hysteresis and long relaxation time. Many structures such as pyramid, hemisphere, pillar, porosity, and other bio-inspired structures have been designed to replace the planar structure to upgrade the device performance. The reduced initial contact area and enlarged structure deformation of sensors enabled by microstructure design result in high compressibility, contributing to higher sensitivity and wider detection range. Following representative examples are shown for illustration of microstructures functioning in performance improvement of flexible pressure sensors.

#### 5.1 | Pyramid structure

Pyramid structure\(^{[73]}\) (Figure 5A) is commonly used in flexible pressure sensors enabling high sensitivity and fast response over low-pressure range.\(^{[217,218]}\) The deformation capability of the devices with pyramid structure can be significantly improved owning to the small shape factor, defined as the ratio of compression area to overall expansion area.\(^{[101]}\) As seen from Figure 5B, the pyramid structured PDMS films perform much higher pressure sensitivity compared with unstructured and line structured PDMS films of the same thickness. Moreover, the structured devices are obtained with superior fast response within hundred milliseconds, while it takes more than 10 s for the unstructured films to return to the original state (Figure 5C). Similar structures\(^{[113–220]}\) have also been adopted to obtain flexible pressure sensors with high performance. For self-powered sensors, microstructures can enhance the triboelectric effect. Furthermore, the self-powered generator with pyramid microstructure exhibited the most excellent performance among line, cubic, and pyramid features. The main reason is that during the friction process, more surface charges can be generated in the complex surface than that in the plane surface. The capacitance change in the deformation process is significantly improved by pyramid microstructures due to the presence of the air voids and the increase in effective dielectric constant.\(^{[73]}\) In addition, the friction charge is easier to separate in the films with pyramid structures, resulting in a larger dipole moment between the electrodes. This research provides a method for the design of self-powered sensors.\(^{[89]}\)

Benefiting from different numerical models developed with varying geometric parameters and materials properties of dielectric layers and lamination layers, the sensitivity of pressure sensors with pyramid arrays is further investigated.\(^{[223–224]}\) As shown in Figure 5D-F, the sensitivity of the sensor is independent on the dielectric constant, and decreases with increasing modulus, which provides guidelines for the material selection of pyramid. To investigate the influence of pyramid geometry on sensitivity, it can be found that the sensitivity decreases with increasing base “a” and increases with upgrading spacing (Figure 5F). Due to the dielectric constant of air lower than that of PDMS, the decrease of pyramid size was demonstrated to be effective for decreasing equivalent dielectric constant and structural modulus, resulting in higher sensitivity of sensors.\(^{[224]}\) Moreover, He et al.\(^{[225]}\) found that the increase of the pyramid height ratio endowed the pressure sensing unit with increased sensitivity but decreased linearity.

And for better understanding the deformation process of pyramid arrays upon compression, the experiments and finite element analysis (FEA) are conducted for compressible pressure sensor (Figure 5G) based on PEDOT:PSS/PUD composite polymer, and the results indicate that the contact surface increases with the pressure.\(^{[101]}\) Figure 5H depicts an integration of a pyramid structured resistive
FIGURE 5  Pyramid structured sensors: design principle and performance. A) Molding of pyramid structure. B) Relationship between capacitance change and pressure of different structured PDMS films. C) Relaxation time of structured films. Adapted with permission.\textsuperscript{[73]} Copyright 2018, Nature Publishing Group. D-F) Influence of structural geometric parameters and material parameters on sensitivity. The inset shows the variation of sensitivity (red) and initial capacitance (green) with the dielectric constant. Adapted with permission.\textsuperscript{[224]} Copyright 2019, Wiley-VCH Verlag. G) Comparison of numerical simulation and experiment on deformation of pyramid structure under compression. Adapted with permission.\textsuperscript{[101]} Copyright 2014, Wiley-VCH Verlag. H-I) Relationship between resistance and pressure at different stages. Adapted with permission.\textsuperscript{[98]} Copyright 2015, Nature Publishing Group. J) Deformation of porous pyramid structure under compression. K-L) Schematic diagram of strain insensitivity of sensor array divided by spacers. Adapted with permission.\textsuperscript{[113]} Copyright 2018, American Chemical Society
pressure sensor and organic electrochromic devices (ECDs). As seen from Figure 5I, the relationship between resistance and pressure is divided into five stages, demonstrating the deformation process of the pyramid structure: in the first stage, the resistance changes slowly, which can be attributed to the deformation mainly concentrating on the tip without SWNT; from the second stage to the fourth stage, the resistance decreases because of the increasing contact area; and in the fifth stage, the contact area does not increase any more, and the resistance changes slowly.

By introducing voids into pyramid structure, the lower compression modulus and larger change of effective dielectric constant can be further enabled in a capacitive pressure sensor based on the porous pyramid dielectric layer (PPDL) (Figure 5J), resulting in the sensing sensitivity up to 44.5 kPa$^{-1}$ within the pressure range of 100 Pa. By placing the pressure sensors on islands of rigid elastomer embedded in a soft substrate with spacers (Figure 5K), the sensing array can be insensitive to in-plane strain (Figure 5L), enabling the measurement in soft curvilinear environment.

### 5.2 Hemisphere structure

Hemisphere structure also plays an enabling role in performance improvement of devices as the contact area of the hemispheres expands continuously during the compression process. Figure 6A shows the schematic diagram of an interlocked structure arranged by two hemisphere structures facing each other. When the sensor was compressed, the hemisphere arrays were squeezed along the normal direction to the contact area. To verify the effects of interlocked structure on pressure sensing, a comparison of the resistance change was conducted with three kinds of structured composite films: a single planar array, a single hemisphere array, and an interlocked hemisphere array. As shown in Figure 6B, the resistance of all the structured films decreases with the applied pressure increasing but with different changing rate. With the applied force, the piezoresistance effect of planar composite films solely relied on the variation of the film resistance ($R_f$) caused by decreased intertube distances in the conductive CNT network. In the single hemisphere structured composite films, the resistance variation included the film resistance ($R_f$) and the contact resistance ($R_c$) between the hemisphere structure and the electrodes, resulting in a greater change than that of planar composite films. For the interlocked hemisphere structure, the change of resistance included the film resistance ($R_f$) of two structured layers and the contact resistance ($R_c$) between the hemisphere arrays, so the variation was greater than that of single hemisphere and planar structured composite films. Thus, the giant tunneling piezoresistance enabled by interlocked hemisphere structure contributed to the high sensitivity and low detection limit (15.1 kPa$^{-1}$, $\sim$0.2 Pa minimum detection). The numerical simulation shows the deformation of the interlocked hemisphere structure under different pressures (Figure 6C), revealing the stress distribution of the contact area and demonstrating the structure to be effective. Furthermore, Park et al. adopted the interlocked hemisphere design to fabricate multimodal electronic skins using flexible ferroelectric films, mimicking the complicated sensing system in human fingertips, in which the multiple spatiotemporal tactile stimuli could be detected and discriminated, such as static and dynamic pressure, and vibration.

Compared with the above structure with the same stiffness of the upper and lower hemispheres, the gradient structure can effectively improve the compressibility and contact area differences, resulting in a larger deformation (Figure 6D). Ha et al. fabricated a flexible triboelectric sensor with two layers respectively made of P(VDF-TrFE) and PDMS. Due to the stiffness gradient structure, the PDMS layer whose elastic modulus was lower than that of P(VDF-TrFE) layer could be more easily deformed. The resulting increasing contact area between the two layers enhanced the triboelectric effect. The influence of hemisphere size was studied numerically. The hemispheres with large size can output large voltage under normal force, while the hemispheres with small size can output large voltage under bending, providing guidelines for structure design of flexible triboelectric sensors adapting to different application scenarios (Figure 6E). Moreover, through introducing nanopores into the interlocked hemispheres, the triboelectric output voltage could be further improved by the increasing gap distance ($\Delta d$) between triboelectric layers according to the Paschen’s law.

The pressure sensors within interlocked structure have been demonstrated to be capable of quantifying different forces and even discriminating the force directions. However, the multisignal detection by comparing the electrical output with the available database leads to time-consuming, and sensing hysteresis. Here, a biomimetic soft e-skin (Figure 6F) based on microstructure can directly discriminate different signals for corresponding forces, including shear force and normal force, suggesting great potentials in the dexterous manipulation of objects in robotics. As shown in Figure 5G, the pyramid and hemisphere structures are arranged facing to each other, and each unit of the e-skin is composed of five capacitive sensors on the cross section. The amplitudes of shear force and normal force could be detected by the corresponding sensor on the top or around the hemisphere. However, two zones in this
design (marked in circles in Figure 6G) have different sensitivities under external force due to the difference of deformation. To achieve high sensitivity for both zone 1 and zone 2, the structure called phyllotaxis spirals inspired from the botany (Figure 6H) is designed, as shown in the bottom of Figure 6I. The position of each pyramid from the center can be given by the formula

$$\phi = n \times 137.5^\circ, r = cn^{1/2}$$

where $n$ represents the numbering order of each single pyramid and $c$ is the scaling parameter. Through the
capacitance distribution diagram in Figure 6J, it can be seen that the device with spiral grid enables the sensor with better sensitivity in the whole region for the force identification. This combination of different structures gives some suggestions to optimizing designs of sensors.

5.3 | Pillar structure

Micropillar arrays have been widely used in the pressure sensors to gain high sensitivity and fast response.[73,230] The uniform geometric size from the bottom to the top of the pillar endows the sensors with better linearity than those with pyramid and hemisphere structures. A piezoresistive pressure sensor based on the micropillar array is shown in Figure 7A.[76] An optimization approach was proposed to improve the pressure sensor with Au micropillar arrays. As can be seen from Figure 7B-C, the pillar diameter is an important factor influencing sensitivity. With increasing pillar size, the sensitivity increases significantly, while the linear region for pressure-resistance decreases tremendously compared with very small and large pillar sizes, that is, 5 and 60 µm in diameter. And owning to the contact resistance affected by pillar size, the pillars of 20 µm diameters enable good linearity for total resistance-contact resistance curve (Figure 7D).

Different from conventional vertical micropillar, a tilted micropillar array is fabricated in dielectric layer to enhance the deformability of a capacitive sensor (Figure 7E).[231] As can be seen from the FEA (Figure 7F), the vertical pillar is compressed and the tilted pillar is bent in the state of compression. And the sensor with tilted structure exhibited larger deformation than that with planar or vertical structure for the same applied pressure, which could be attributed to the fact that the bending strain energy is much smaller than the compressive strain energy. The sensing performance was also influenced by the geometry of tilted micropillar. For instance, the capacitance response could be enhanced by the increase of the length width ratio of the micropillar. Additionally, the angle of tilting and the ratio of the micropillar diameter to the center-to-center distance should be taken into account as well, which could affect the sensitivity, stability, and recovery time of the sensor.

Inspired from the merkell cell papillae structure, the i-TPU pillar microstructure (Figure 7G) is introduced on viscopore elastic membrane, which effectively increases the output stress and induces the effective extrusion of ionic liquid under low applied force.[232] As shown in Figure 7H, under the same conditions of pressure and voltage, the capacitance change of the film with i-TPU pillar microstructure is greater than that of flat film. The FEA further indicates that pillar structure cause more stress than flat structure when they are subjected to the same loading, resulting in the high sensitivity of pillar structured pressure sensors under low stress stimuli (Figure 7I).

By exploiting hierarchical morphologies, the performance of micropillar structured sensors can be further enhanced. As shown in Figure 7J, two types of sensors based on the structure with hierarchical ZnO NWs fabricated on the PDMS micropillar arrays, are capable of sensitively detecting static and dynamic tactile stimuli respectively based on piezoresistive effect and piezoelectric effect.[233] The hierarchical structures here coated with metal could induce a great increase in the surface area, resulting in a sharp decrease in contact resistance under static pressure. As can be seen from Figure 7K, the sensitivity of the sensor with pitch are higher than that with planar micropillar, and the sensitivity increases with the decrease of the pitch size. Simultaneously, the planar PDMS suffers from the thermal expansion over 30–90°C while the hierarchical ZnO nanowire (NW) arrays render the temperature-insensitive capability of the sensor, enabling a stable pressure sensitivity (Figure 7L). The upper layer of ZnO NWs was coated with Ni films, and the bottom layer was comprised of bare ZnO NWs. When dynamic pressure is applied, the bare NWs with low stiffness bend under the press of the NWs coated with Ni, producing positive potential on the extended end and negative potential on the compressed end due to the Schottky barrier between the Ni and the ZnO (Figure 7M). Owning to the small size of nanowires, the piezoelectric sensor could detect stimuli up to 250 Hz.

5.4 | Porous structure

Pyramid, hemisphere and pillar structured sensors exhibit higher sensitivity than planar structured sensors, benefitting from the smaller initial loading area and larger deformation during compression. Some researchers worked on another way to improve the sensing sensitivity,[234–237] that is, introducing porous structures into materials, which can be used to decrease the modulus of the materials effectively.

PPy as a kind of commonly used conductive polymers in flexible pressure sensors can be obtained with lower modulus after being made into a porous structure with millimeter-sized voids. And through a multiphase reaction, the micrometer-sized voids can be fabricated in PPy hydrogel film, endowing the PPy with better elasticity and lower elastic modulus (Figure 8A, B) than that with millimeter-sized voids to satisfy the requirement of flexible pressure sensors.[94] Thereby, its corresponding sensor was more sensitive than the resistive sensor employing PPy-coated PE foam,[238] and was 25 times more sensitive and more faster in response than the capacitive sensor.
based on polyolefin foam. Additionally, according to the results of compression experiments, the elastic modulus of the porous PPy film approached that of PDMS when the pressure was 5 kPa, demonstrating the good flexibility of the PPy porous structure. Also, due to the existence of voids, the normalized density modulus of PPy was lower than that of a polymer doped with CNTs, and the hysteresis of PPy under low pressure was also significantly lower than that of other conductive polymers. The above comparisons suggest that the voids significantly improve the properties of PPy and show the great potential of porous PPy in the field of flexible electronics.

By controlling pore morphology and designing patterned arrays, the influence of different pore size for devices has been further studied. According to the FEA (Figure 8C), the smaller modulus achieved by the...
FIGURE 8  Porous structured sensors: design principle and performance. A) Schematic of deformation of porous structured PPy under compression. B) SEM of PPy hydrogels synthesized with porous microstructures. Adapted with permission. \(^{[94]}\) Copyright 2014, Nature Publishing Group. C) Finite element analysis of porous rubbers with different pore size. Adapted with permission. \(^{[77]}\) Copyright 2014, Wiley-VCH Verlag. D) Schematic of the simplified model for the porous structure through pillars with a applied force. E) Relationship of different structured pressure sensors between capacitance change rate and pressure. Adapted with permission. \(^{[240]}\) Copyright 2016, Wiley-VCH Verlag. F) E-skin with porous PDMS and air gap capable of discriminating multiple mechanical stimuli. G) Relationship of different structured pressure sensors between capacitance change rate and pressure. Adapted with permission. \(^{[91]}\) Copyright 2014, Wiley-VCH Verlag. H) Schematic of fluorocarbon piezoelectret pressure sensor. I) Potential distribution of the FPS under compression. J) Transferred surface charge density increasing with applied pressure. Adapted with permission. \(^{[122]}\) Copyright 2017, Elsevier BV. K) Schematic of bean pod-like structured sensor and response curves of pressure sensors with different size PS microspheres (3, 1.3, and 0.8 µm). L) SEM images of the surface with different size PS microspheres (i) 0.8 µm, (ii) 1.3 µm, and (iii) 3µm. Adapted with permission. \(^{[59]}\) Copyright 2020, American Chemical Society. M) Hierarchical NP-MP structure in mechanochromic composite. N) Relationship between normal force and light intensity of different pore sizes and different SNP sizes with 5 µm pores. Adapted with permission. \(^{[260]}\) Copyright 2019, Wiley-VCH Verlag
bigger pores results in larger overall deformation, contributing to larger resistance variation (i.e., higher sensitivity). Lee et al.\cite{240} studied the influence of pore size on the capacitive pressure sensors. Figure 8D shows that the relative capacitance change of the porous structured PDMS is five to eight times larger than that of the bare PDMS. The impressive enhancement in sensitivity of sensors with pores could be attributed to two factors that were the pores making the structure softer, and the increase of dielectric constant. It can also be concluded from Figure 8E that the sensitivity can be upgraded with the increase of pore size. The influence of pore sizes on deformation is studied by simplifying the unit support to an isotropic elastic half pillar, whose stiffness can be solved by the formula\cite{241}

\[
S = \frac{2 E \eta (r + r_c)}{(1 - \nu^2)},
\]

where \(\nu, E\) are the Poisson’s ratio and shear modulus of the half-space respectively, \(\eta\) is a constant chosen to account for the more complex geometry of the postbase connection. The \(r\) and \(r_c\) represent the pillar radius and the fillet radius of the structure. Based on the equation, the compression ratio under a constant pressure could be theoretically calculated according to the pore sizes of the films. Both of the theory and the experimental research indicated that the larger pore sizes could upgrade the compressibility of porous structured films, leading to a higher sensitivity.

Combining porous PDMS structure and air gap, Park et al.\cite{91} proposed a stretchable energy harvesting e-skin (EHES) that could discriminate, and harvest various mechanical stimuli, such as compression, stretching, and bending. As shown in Figure 8F, the thin films of SWNTs are utilized as top and bottom electrodes while the porous PDMS structure and air gap are adopted as dielectric layers. The normal force and compression could be detected with the capacitance, and the lateral deformation due to tension or bending was measured by the resistance, such that the different mechanical stimuli could be detected and discriminated. Under a normal force, a large increase in capacitance was observed, with negligible change in the resistance. Figure 8G shows that the porous structure and the air layer improve the sensitivity of the sensor in low-pressure region (<1 kPa), and high pressure region (>1 kPa), respectively, which enables a wide pressure detecting scope. Moreover, the power of the device was generated by the friction between PDMS and SWNT layers. The design of this sensor opens up the direction for multifunctional sensors.

Compared with porous PDMS films, the porous piezoelectret polymeric materials with space charge exhibit high thermal stability, superior piezoelectric property, and good flexibility. The size of the air voids is an important factor whose increment will reduce the Young’s modulus and greatly enhance the piezoelectric coefficient. Many methods have been developed to increase the void size, such as pressure expansion,\cite{242–244} double-expansion process\cite{245} and hot-pressing.\cite{246} A capacitive pressure sensor (FPS) developed by Zhou et al.\cite{212} based on FEP, FEP/F-PTFE films is shown in Figure 8H with the working mechanism illustrated in Figure 8I. As shown in Figure 8J, the large voids enable a high sensitivity up to 7380 pC/N in the subtle-pressure regime (<1 kPa), which suggests good performance of piezoelectric electret. Xiaoqing Zhang et al. fabricated piezoelectric self-powered sensors by using solid PTFE and porous PTFE laminated, and the power generation efficiency of the porous PTFE can also be improved by increasing the hole size.\cite{247}

Porous structures function well in performance enhancement of pressure sensors but they are still suffering from poor resilience. Aiming to sustain more complex mechanical stresses, hierarchical structures over a nanoscale have been demonstrated to be effective.\cite{248,249} Thus, a bean-pod like structure was proposed to achieve the coupling of different structures over a multiscale.\cite{59} In such structure, the large internal cavity served as a storage for elastic material, thereby improving the rebound characteristics and structure durability. As seen from Figure 8K, a resistive sensor with bean-pod like structure is assembled by two laser-induced graphene/polyurethane (LIG/PU) films facing to each other and the cavities of the graphene are filled with elastic polystyrene (PS) microspheres.\cite{59} Compared with the structure without PS microspheres, PS filled porous graphene structure could improve the electrical conductivity. And the relationship between the size of PS microspheres and the sensitivity of sensor was further investigated. Among the three size of experimental study, the microsphere with the diameter of 1.3 \(\mu\)m performs the highest sensitivity (Figure 8K). Seen from the scanning electron microscopy (SEM) (Figure 8L (i-iii)), the microspheres of 0.8 \(\mu\)m almost fall within the LIG pores, unable to protrude beyond the LIG cavity to effectively form a space layer, resulting in no significant sensitivity improvement; the 1.3 \(\mu\)m microspheres successfully protrude outside the LIG cavities, enabling the microsphere spacer to effectively tune the generation of new contact points and the contact area change between the LIG layers during compression, contributing to a higher sensitivity; for the 3 \(\mu\)m microspheres, the microspheres fail to work as an insulating spacer layer limited by the low quantity although the protrusion is achieved. As shown in Figure 8M, another hierarchical structure is composed of spiropyran (SP), poly(dimethylsiloxane) (PDMS), and silica nanoparticles (SNPs).\cite{250} SNPs with nanoscale diameter induced stress...
concentration in micropore-based PDMS and improved the mechanochromic sensitivity SP material. As can be seen from the comparison in Figure 8N, with the reduction of the micropore size, the transition pressure of mechanochromic material decreases from 2.5 to 1.8 N, and the addition of the nano-fillers further reduces the transition pressure, resulting in higher sensitivity, and the transition pressure of the material can be lowered to 1 N. The hierarchical nanoparticle-in-micropore (NP-MP) was demonstrated to be a novel and effective preparation strategy for flexible pressure sensors.

In the above introduction, the flexible pressure sensors with four kinds of microstructures are analyzed from the geometry and the materials. The performance of microstructured sensors based on different materials is listed in Table 1, including sensitivities, LOD, response time, and relaxation time. It may hopefully provide a reference for the microstructure design of sensors.

5.5 Other structures

In addition to the pyramid, hemisphere, pillar, and porous structures, there are many other forms of strategic microstructures, which can effectively improve the performance of the pressure sensor. Several representative works are shown as following.

The sensors with nano-needle arrays (Figure 9A) can be fabricated by Breath figure method.[259] And compared with nonstructured and hemispherical structured sensors, the sensors with needle structure exhibits a much higher relative capacitance change rate (Figure 9B). Mathematical models also were established to further investigate the mechanics of this structure, and the results indicated that the displacement of the structure was inversely proportional to the actual loading area under a certain pressure, enabling the needle structure to perform high sensitivity.

Three-dimensional (3D) carbon nanofiber networks (CNFNs) (Figure 9C) inspired from human body hair have been proposed with superior pressure-sensitivity through electrospinning and thermal treatment.[260] Based on the porous nanostructure, the sensitivity of pressure sensor reached 1.41 kPa⁻¹. And due to the nanoreinforce of Al₂O₃, the CNFNs exhibited higher flexibility, resilience, and compressibility than traditional carbon-based sensors. Besides the normal pressure detections, the directions of shear forces can be successfully discriminated through the specially designed arch-array (Figure 9D).

Lotus leaf surface is a natural multiscale hierarchical structure (Figure 9E).[275] The graphene/PDMS structure designed by imitating the structure makes the sensor obtain good sensitivity. The numerical results show that the multiscale structure has a larger change of contact area than the smooth structure (Figure 9F), and the corresponding sensitivity is higher under low pressure. However, with the increase of graphene thickness, the initial resistance and sensitivity of the sensor decreased, demonstrating the graphene of proper thickness was the key to this multiscale structure design.

Some flexible pressure sensors have achieved the ability of force positions discriminating in the nonstretched state,[261,262] but the staircase structural design even functions under stretching. A novel tactile electronic skin sensor assembled back-to-back by two antiparallel stairs based on vertically arranged v-AuNWs is presented in Figure 9G.[263] With an increase in strain level from 0% to 50%, the overall resistance of the staircase film was increase owning to the strain-induced cracks appearing in each step of the film. But the sensitivity ratio of sensors 1 and 2 (S₁/s₁/2), namely, the location-specific dimensionless parameter L₁, from location 1 to location VI remained unchanged with different applied pressures. And the correlation of predicted and experimentally measured L₁ subjected to pressure ranging from 4 to 40 kPa is demonstrated in Figure 9H, showing that the sensor can predict precise position even with a tensile.

Pyramid, pillar, and hemisphere structured pressure sensors exhibit good sensitivity in the small pressure range, but the sensitivity decreases with increasing pressure. On the other hand, nanowire structure enables the pressure sensors a uniform geometry and good linearity, but the sensitivity is relatively low. Compared with these conventional microstructures, a special surface topography with random distribution spinosum (RDS) microstructure can achieve large sensitivity and good linearity simultaneously in a wide pressure range, due to its strong structural hierarchy, as shown in Figure 9I, J.[264] The results of numerical simulation show that the stress concentration appears at the top of the structure under low-pressure loading, the tip structure is compressed first, showing a high sensitivity. With the deformation of the structure, new contacting points are formed, thereby reducing the overall resistance and maintaining high sensitivity. The new contact points and the expanding contact area maintain the large sensitivity and linearity in a large pressure range. In addition to the influence by microstructures, sensitivity and measurement range are also affected by the strain hardening of polymer materials, where sensitivity decreases with the increase of pressure. The key to solve this problem is to develop highly sensitive materials and strategic microstructures simultaneously. N. Bai et al. fabricated iontronic pressure sensor based on graded intrafillable architecture (GIA) with sandpaper, which showed high sensitivity coefficient in a wide pressure range.[277] Compared with hemispherical or inclined cylindrical structure, the GIA structure is easier to deform,
FIGURE 9  Other structured sensors: design principle and performance. A) Schematic of the capacitance pressure sensor with nano-needle PU film. B) Relationship of different structures between relative capacitance change and pressure. Adapted with permission.[259] Copyright 2011, Springer Verlag. C) Schematic of the sensor with 3D carbon nanofiber networks (CNFNs) inspired from human body hair. D) Asymmetric structure of the arch CNFN unit. Adapted with permission.[260] Copyright 2019, Royal Society of Chemistry. E) Pressure sensor based on lotus leaf surface structure. F) Numerical simulation of hemispheres with different surface structures under pressure. Adapted with permission.[275] Copyright 2019, Wiley-VCH Verlag. G) Schematic of a location-specific sensor with a uniaxial strain of 50%. H) The correlation of predicted and experimentally measured Ln subjected to pressure ranging from 4 kPa to 40 kPa. Adapted with permission.[263] Copyright 2018, Royal Society of Chemistry. I) Finite element analysis for different geometries: RDS, hemisphere, pyramid, nanowire, at an external loading pressure of 5 kPa. J) Resistance variation versus applied pressure for different surface microstructures. Adapted with permission.[264] Copyright 2018, American Chemical Society
| Mechanism   | Material                  | Sensitivity 1, Range, Linearity | Sensitivity 2, Range | LOD       | Response Time | Relaxation time | Ref. |
|------------|---------------------------|---------------------------------|----------------------|-----------|---------------|-----------------|------|
| Pyramid    |                           |                                 |                      |           |               |                 |      |
| Piezoresistance | PDMS/Graphene        | -5.53 kPa·m⁻¹, <100 Pa         | -0.1 kPa·m⁻¹, >100 Pa | 1.5 Pa   | 0.2 ms        |                  | [82] |
| Piezoresistance | PDMS/Au              | 6.7 × 10⁷ kPa·m⁻¹, 1–5 kPa    | 3.8 × 10² kPa·m⁻¹, 5–50 kPa |         |               |                  | [28] |
| Piezoresistance | PDMS/PEDOT:PSS       | 4.88 kPa·m⁻¹, 0.37–5.9 kPa    | 93 mg                |           |               |                  | [101]|
| Piezoresistance | PDMS/Au NWs          | 23 kPa·m⁻¹, <600 Pa           | 0.2 Pa               | <10 ms   | 30 ms         |                  | [113]|
| Capacity   | PDMS/ITO              | 44.5 kPa·m⁻¹, <100 Pa         | 0.14 Pa              |           | 100 ms        |                  | [111] |
| Capacity   | MPIG/ITO/PET          | 41 kPa·m⁻¹, <400 Pa           | 4 Pa                 | <20 ms   |               |                  | [219] |
| Capacity/OTFT | PDMS/PET/ITO         | 0.55 kPa·m⁻¹, <0.2 kPa        | 3 Pa                 | <1 s     |               |                  | [74]  |
| Triboelectricity | PDMS/Ag NWs       | 0.31 kPa·m⁻¹                   | 2.1 Pa               | <5 s     | 5 ms          |                  | [140] |
| Hemisphere |                           |                                 |                      |           |               |                 |      |
| Piezoresistance | PDMS/Graphene       | 110 kPa·m⁻¹, 0–0.2 kPa        | 3 kPa·m⁻¹, 0.2–15 kPa | 0.2 Pa   | 30 ms         |                  | [231] |
| Piezoresistance | PDMS/PS              | -15 kPa·m⁻¹, <100 Pa          | 4 Pa                 | <100 ms  |               |                  | [83]  |
| Piezoresistance | PDMS/PS              | 196 kPa·m⁻¹, <5 kPa, 98.37%   | 0.5 Pa               | <26 ms   |               |                  | [252] |
| Piezoresistance | PDMS/CNTs           | 15.1 kPa·m⁻¹, <0.5 kPa        | 0.2 Pa               |           |               |                  | [75]  |
| Triboelectricity | PDMS/PVDF          | 0.55 V/kPa                    | 1 Pa                 | <1 s     |               |                  | [226] |
| Pillar     |                           |                                 |                      |           |               |                 |      |
| Piezoresistance | PPy/PDMS            | 1.80 kPa·m⁻¹, <0.35 kPa       | 0.03–17 kPa·m⁻¹, <1 kPa | 2 Pa     |               |                  | [76]  |
| Piezoresistance | PDMS/Au             | 2.0 kPa·m⁻¹, 0–0.22 kPa       | 0.87 kPa·m⁻¹, 1.0–3.5 kPa | 15 Pa    | 50 ms         |                  | [253] |
| Piezoresistance | PDMS/ZnO            | -6.8 kPa·m⁻¹, <0.3 kPa        | 0.6 Pa               | <5 s     |               |                  | [39]  |
| Piezoresistance | PDMS/Ag NWs         | 20.08 kPa·m⁻¹, 0.05–0.8 kPa  | 3.81 kPa·m⁻¹, 0.8–2.1 kPa | 50 Pa    |               |                  | [239] |
| Porosity   |                           |                                 |                      |           |               |                 |      |
| Piezoresistance | PPy                  | 7.7–41.9 kPa·m⁻¹, <100 Pa     | <0.4 kPa·m⁻¹, >1 kPa  | <1 Pa    |               |                  | [94]  |
| Piezoresistance | PDMS/Graphene       | 15.9 kPa·m⁻¹, 60 kPa          |                     |           | 1.2 ms        |                  | [254] |
| Piezoresistance | PLA/MXene           | 0.55 kPa·m⁻¹, 23–982 Pa       | 3.81 kPa·m⁻¹, 0.982 -10 kPa | 10.2 Pa  | 11 ms         | 25 ms           | [285] |
| Piezoresistance | PDMS/Wood           | 10.74 kPa·m⁻¹, <100 kPa, 99% |                     |           | 20 ms         | 20 ms           | [256] |
| Capacity   | PDMS/ITO             | 0.26 kPa·m⁻¹, 0–0.33 kPa      | 0.01 kPa·m⁻¹, 0.33–250 kPa |           |               |                  | [165] |
| Capacity   | Ecoflex/CNTs         | 0.601 kPa·m⁻¹, <5 kPa         | 0.077 kPa·m⁻¹, 30–130 kPa | 0.1–0.2 Pa |               |                  | [257] |
| Piezoelectric/OFET | Cellular PP        | 0.1 V/kPa, <400 kPa          |                     |           | 2 kPa         |                  | [211] |
| Piezoelectric | THV/COC              | 0.03 V/kPa, <150 kPa, 99%    |                     | 150 kPa  | ~1.5 ms       |                  | [288] |

because the undercuts and grooves on the surface offer space for surrounding structures to accommodate deformation. Additionally, under pressure, the contact area between GIA structures and electrode changes much more rapidly than other structures. Moreover, as the GIA structures were fabricated by ionic electronic materials, such an increase in contact area resulting in large capacitance change, which is 5–6 orders of magnitude higher than that of the noniontronic materials. For these two reasons, the sensor can achieve high recognition accuracy for light objects under high pressure. This method provided another solution to the dilemma between good sensitivity and large measurement range.

The nature endows organisms with the best structure to adapt to the environment. Some flexible pressure sensors based on biomimetic microstructure are listed in Table 2.
| Structure               | Mechanism            | Material         | Sensitivity 1, Range, Linearity | Sensitivity 2, Range, Linearity | LOD | Response Time | Relaxation time | Ref. |
|-------------------------|----------------------|------------------|-------------------------------|---------------------------------|-----|---------------|----------------|------|
| Sand Paper Mimicking    | Piezoresistance      | PDMS/Graphene    | 2.5 kPa⁻¹, 0.01–1 kPa         | 12.0 kPa⁻¹, 1–50 kPa            | 10 Pa | <120 ms       | 80 ms          | [265]|
| Epidermis Mimicking     | Piezoresistance      | PDMS/Graphene    | 25.1 kPa⁻¹, 0–2.6 kPa         | 0.45 kPa⁻¹, >2.6 kPa            | 16 Pa | <150 ms       | 40 ms          | [264]|
| Wave                    | Piezoresistance      | PDMS/CNTs        | 0.152 kPa⁻¹, 0–3.24 kPa       | 0.0049 kPa⁻¹, 12–27 kPa         | ~162 ms | 108 ms       |                | [266]|
| Wave                    | Capacity             | PDMS/Ag NWs      | 2.04±0.16 kPa⁻¹, 0–2 kPa      | 0.57±0.08 kPa⁻¹, 2–9 kPa        | <7 Pa | <100 ms       | 100 ms         | [267]|
| Wrinkle                 | Piezoresistance      | AAO/Graphene     | 6.92 kPa⁻¹, 0.3–1.5 kPa       | 0.14 kPa⁻¹, 1.5–4.5 kPa         | 300 Pa |                |                | [268]|
| Buckled                 | Capacity             | PDMS/Ag NWs      | 3.8 kPa⁻¹, 45–500 Pa          | 0.8 kPa⁻¹, 2.5 kPa              | 45 Pa | <30 ms        | 60 ms          | [111]|
| Natural Petal           | Piezoresistance      | PDMS/Graphene/CNTs | 19.8 kPa⁻¹, <0.3 kPa       | 0.27 kPa⁻¹, 1–6 kPa            | 0.6 Pa | 16.7 ms       |                | [269]|
| Mesh                    | Piezoresistance      | Carbonized Silk  | 34.47 kPa⁻¹, 0.8–400 Pa       | 1.16 kPa⁻¹, 0.4–50 kPa         | 0.8 Pa | 16.7 ms       |                | [270]|
| Network                 | Piezoresistance      | PDMS/TPU         | 5.54 kPa⁻¹, <10 kPa          | 0.123 kPa⁻¹, 10–100 kPa        | ~10 Pa | ~20 ms        | 30 ms          | [71] |
| Spherulite              | Piezoresistance      | MP-ZnO/PS        | 10 kPa⁻¹, 0–2 kPa            | 3.4 kPa⁻¹, 7–11 kPa            | 14 ms |                |                | [271]|
| Needle                  | Piezoresistance      | Dried Flower Petal | 1.54 kPa⁻¹, 0.6–115 Pa     |                              |      |                |                | [272]|
| Lotus Leaf Mimicking    | Capacity             | PDMS/Ag NWs      | 1.94 kPa⁻¹, <2 kPa          | 0.077 kPa⁻¹, >2 kPa            | <0.8 Pa | 36 ms        | 58 ms          | [273]|
| Lotus Leaf Mimicking    | Capacity             | PDMS/PS/Au       | 0.815 kPa⁻¹, <0.1 Pa        | 0.0047 kPa⁻¹, >100 Pa          | 17.5 Pa | ~38 ms        |                | [274]|
| Lotus Leaf Mimicking    | Piezoresistance      | PDMS/Graphene    | 1.2 kPa⁻¹, 0–25 kPa         |                              | 5 Pa |                |                | [275]|
| Nanowire                | Piezoelectricity     | PbTiO₃           | 9.4 × 10⁻³ kPa⁻¹         |                              | 5 ms | 7 ms          |                | [88] |
| Nanowire                | Piezoelectricity/OTFT| PDMS/ZnO         | 21 ms kPa⁻¹, <3.5 kPa       |                              |      |                |                | [86] |
| Nanowire                | Triboelectricity     | PET/FEP          | 44 mV Pa⁻¹, <0.15 kPa, 99%  | 0.5 mV Pa⁻¹, >2 kPa, 97%       |      |                |                | [136]|
| Nanowire                | Triboelectricity     | PTFE-Nylon       | 51 mV Pa⁻¹                  |                              |      |                |                | [276]|
| Graded Intrafillable    | Capacity             | PVA/H₂PO₄        | 220–3300 kPa⁻¹, 0.08 Pa–360 kPa |                              | 0.08 Pa | 9 ms        | 18 ms          | [277]|

TABLE 2 Parameter comparison of other structured pressure sensors
It can be seen from the table that these sensors have good performance.

6 | APPLICATION

Benefitting from all the aforementioned achievements, the microstructured flexible pressure sensors significantly boost great advances in various intelligent systems, involving health monitoring, e-skin, robotics, human–machine interface, etc. Here, some representative examples in these areas are highlighted.

Flexible pressure sensors enabled by microstructure can detect very subtle texture change, such as respiration, blood pressure, heart beats, boosting the usage in mobile health monitoring and remote diagnostics in some chronic diseases and rehabilitation monitoring.\(^{[73,99,100,271]}\) And the hierarchical structure of combing several different microstructures, can further improve device performance. As seen from Figure 10A,B, a combination of microhairy and pyramid structures renders the pulse signal amplification, due to the enhanced adhesion by microhair and improved sensitivity by pyramid, with distinguishing advantages for diagnosis of cardiovascular and cardiac illnesses.\(^{[217]}\) The Schottky diodes is formed by using PEDOT:PSS microspheres particles (MPs) as cathode and ZnO/PS composite as anode on the irregular surface (Figure 10C).\(^{[278]}\) The Schottky diode structure enhanced by MPs contributed to eliminating electrical crosstalk of the sensors, which enabled the sensor to achieve high resolution for Braille reading as an artificial fingertip.

Taking advantage of the microstructure, the polymer-based sensors are also capable of detecting dynamic pressure as the response speed can be improved with little influence from the intrinsic viscoelastic behavior of materials, suggesting the applications in some high-frequency environments. Inspired by the human eardrum, a self-powered bionic membrane (BMS) is shown in Figure 10D fabricated by Son et al.\(^{[282]}\) through combining the contact electrical effect of PTFE and nylon with the eardrum structure. In order to enhance the triboelectricity, the vertically aligned polymer nanowires were created onto the PTFE surface, improving the effective contact area between the nanowires of PTFE, as well as the output performance. The oval PTFE tympanic membrane became tense due to the fixed periphery, exhibiting fast response to dynamic pressure over a wide frequency range, even capable of detecting the throat sound at high frequencies up to 1500 Hz. With the pyramid microstructured triboelectric friction layer and electrolyte (Figure 10E), a newly designed pressure sensor is fabricated based on the potentiometric-triбоelectric hybridized sensing principle (Figure 10F), capable of detecting static and dynamic pressure through a single mode, replacing of one platform comprised of different sensing elements and multimode sensing devices based on the combination of different sensing principles.\(^{[279]}\) The design method of this sensor and the decomposition method of electrical signals provide a new clue for designing multifunctional sensors.

The flexible pressure sensors conformal to joints can discriminate different motions, such as bending of finger, wrist and elbow,\(^{[118,219,266,272]}\) facilitating some applications of human–machine interface. Figure 10H shows a capacitive sensor with dry rose petals as the dielectric layer, which can be integrated into the glove to accurately recognize the stretching or bending gesture of fingers (Figure 10H-J).\(^{[272]}\)

As pressure sensors can transfer the outside stimulus into a signal, they are capable of functioning in the AI systems to actuate a behavior of an object, such as grasp, movement, jump, etc.\(^{[218,254]}\) As shown in Figure 10K, the pyramid structured pressure sensors are integrated into a flexible artificial afferent nerve developed by Kim et al.\(^{[96]}\) By adjusting the size of the micropyramid, the pressure sensor had the same working range with high sensitivity as the biological receptor and adapted to the reaction characteristics of the organism. And the pressure information collected from clusters of pressure sensors could be converted into action potentials via ring oscillators. The pressure sensors embedded artificial afferent nerve could motor nerves to actuate muscles through building up a hybrid bioelectronic reflex arc, with great potential application in neuro-robotics and neuroprosthetics.\(^{[280,281]}\)

Due to the soft and highly integrated requirements of flexible electronic devices, multifunctional sensing is the inevitable trend of flexible sensors.\(^{[9,282,283]}\) Great progress has been made in the measurement of multiple parameters simultaneously using a single sensor.\(^{[35,284]}\) Park measured the dynamic pressure and temperature by the piezoelectric and thermoelectric effects of PVDF/reduced graphene oxide (RGO) composites, and measured the static stress by the change of contact resistance.\(^{[99]}\) The spherical microstructure design of contact surface improves the contact resistance of the sensor, and endows the sensor with high accuracy in the measurement of light objects such as water drops. Levent Beker proposed a two-layer sensing method, which combines membrane structure with pyramid structure.\(^{[218]}\) The first layer is composed of a stretchable film on which resistance or capacitance sensors are fabricated to detect the surface of the contact object. The pyramid pressure sensor in the second layer can measure the applied force on the object to avoid over-loading. Although these sensors can measure many categories of parameters, the human skin are far beyond these advanced multifunctional sensors in sensing parameter...
diversity. Therefore, the integration of bio-inspired sensing functions through structure design and material modification is a significant segment of research interest for multifunctional sensor.

7 | CONCLUSION

In this paper, the development of flexible pressure sensors in recent years is systematically summarized. The
working mechanisms of sensors are briefly described. The flexible pressure sensors are divided into functional layer, electrode layer and substrate layer according to the function of the layer with corresponding materials. Microstructures through micro/nano processing technology can effectively improve the performance of flexible sensors. The pressure sensors with pyramid, hemisphere, pillar, porous and other structures are introduced in detail. The sensitivity, LOD, measurement range, response time and other performances of various sensors can be clearly compared from the Table 1 and Table 2, demonstrating the superiority of the microstructured sensors. Based on parameter study and numerical simulation, some suggestions and guidelines are carried out for optimizing structure design.

Although pressure sensors can be enabled with better performance through the microstructure design, but most works on them are still in the laboratory stage, far away from practical use. The pyramid structured pressure sensors exhibit high sensitivity and linearity in the low-pressure range, while their linearity range is too limited. The hemisphere structured pressure sensors highly rely on the contact area change which is uneven and whose linearity is poor during the compression process. Benefiting from the uniform structure, the pillar structured pressure sensors perform good linearity but the pressure sensitivity is relatively low. The porous structure endows sensors with lower modulus and better flexibility but limited by the preparation technology of porosity, the distribution of voids in the structure cannot be uniform, resulting in poor consistency of sensors. Biomaterials with natural structure inside have been used in some sensors but due to their tissue specificity and poor stability, they are not suitable for direct use in the preparation of sensors. Some bioinspired structured sensors have been developed but their geometry characteristics and material properties have not been studied deeply. As for the available preparation techniques, they are too complicated for microstructure fabrication to reach mass production.

Therefore, we believe that the flexible pressure sensors can be further studied from improving the measurement accuracy, response speed and self-adapting to sophisticated application scenarios. Based on expanding the linear measurement range of the sensor through innovative structures or materials, the measurement accuracy of the sensor can be enhanced. Establishing accurate calibration methods for sensors (i.e., pressure–electricity nonlinear relationship) is another effective approach for higher measurement accuracy. By improving the material and optimizing the structure, the hysteresis caused by the viscoelasticity of polymer materials may be reduced towards the practical application of the flexible pressure sensor. Interference factors (e.g., temperature, humidity, magnetic field) in a complex environment cannot be inevitable for practical use. For instance, polymer materials may suffer from mechanical behavior change by temperature variation and thermal expansion by temperature rising, resulting in electrical signal change. It is desirable to establish the calibration method with taking account of various interference factors.

Above all, considerable efforts are expected to be made for practical application of pressure sensors, with solving several key scientific problems and developing innovative techniques.

ACKNOWLEDGMENTS
The authors acknowledge the National Key Research and Development Program of China (no. 2020YFC2007101), National Natural Science Foundation of China (nos. 61933002 and 11972325), Natural Science Foundation of Zhejiang Province of China (nos. LQ20A020005 and LQ19E030003).

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**How to cite this article:** Tang R, Lu F, Liu L, et al. Flexible pressure sensors with microstructures. *Nano Select.* 2021;1-28. https://doi.org/10.1002/nano.202100003.