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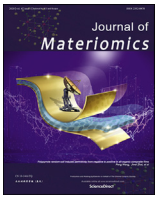

Recent advances of wearable and flexible piezoresistivity pressure sensor devices and its future prospects

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ABSTRACT

The human skin inspired soft electronic devices have attracted broadly research attention in the past decades as the promising potential applications in health monitoring and diagnosis, robotics, and prosthetics. The soft wearable piezoresistivity pressure sensor is one of the most attractive candidates for the development of advanced electronic skin for its simple mechanism, compact structure, low cost and power energy consumption and ease of signal acquisition and transforms advantages. In this review, we will explore the recent progress and achievements in the field of piezoresistivity pressure sensor, focusing on the fundamentals of the piezoresistivity pressure sensor and the materials related to the devices, including active materials, substrate materials, and electrode materials. Subsequently, the challenges and outlook are discussed. We list several current challenges perspectives on the development of pressure sensors. Several critical topics for the optimization of the sensitivity and working range of sensing devices toward practical applications are discussed. Finally, perspectives on the slip and force vectors sensors, the developing technologies for multi-function and high-resolution sensor systems and signals process technologies are examined to highlight the near future development tendency in piezoresistivity pressure sensor research field.

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1. Introduction

Flexible and wearable devices that present the properties of soft human skin, such as tactile sensing capability, toughness, self-healable and stretchability, have attracted much attention in the past decades with the development of mobile devices, robotics, and prosthetics [1–12]. As the next generation of electronic products, wearable soft devices can be set to merge with our bodies to extend our perceptions or provide vital insights into the health of individuals in real-time. Benefit from its prominent sensing features, it can act as the artificial skin or electronic skin (e-skin) for the biomimetic prosthesis and robot, which would greatly improve the human-machine interaction and promote the advancement of artificial intelligence systems.

Pressure sensing ability is one of the basic yet powerful features of human skin. The human can effortlessly distinguish ultralow pressure even generated by a light breeze. Furthermore, our powerful pressure sensing ability also helps us to distinguish the roughness, softness, weight, and shape of objects. These multidimensional tactile feedback information help us better control of our forces. There are four types of mechanoreceptors in human skin. These receptors can measures forces with different receptive field sizes or on different time scales, thus our skin can response to static and dynamic forces with high sensitivity and resolution. Furthermore, the soft mechanical properties of the skin can help skin surface adapts to the surface of an object and providing more contact area for sensory perception, which can improve its sensibility.

The soft organic polymer-based flexible and stretchable pressure sensors provide a novel potential opportunity to mimic the sensing and soft mechanical properties of human skin. In recent years, great efforts and achievements have been demonstrated in pursuing artificial electronic skin. Various kinds of pressure sensors have been developed with different structures and transduction mechanisms, such as capacitance, piezoresistivity, and piezoelectricity. Among these devices, piezoresistive sensors, which transduce pressure stimulate into a resistive signal, have been widely used for its simple sensing mechanism, ease of constructing, low power consumption advantages. In the following sections, we briefly summarize the main discussions and the motivation underlying each topic of this review. In section 2, we attempt to summarize the mechanism and structure of pressure sensors; In the next section, we mainly focus on the development of materials, including, active materials, substrate materials, and electrode materials, for the designing and fabrication of device; Then, the challenges and outlook are discussed in section 4. We list several current challenges perspectives on the development of pressure sensors: First, advances and challenges of E-textile pressure sensor devices toward practical applications are discussed; Second, specifically issues about slip and force vectors sensors, integration and fabrication technologies for multi-function and high-resolution sensor system, power consumption and signals process technologies are explored.

2. Fundamentals of piezoresistivity pressure sensors

2.1. Transduction mechanism and structure of pressure sensors

Piezoresistive sensors transduce the resistance electrical signal change of a device into an applied pressure. The structure of piezoresistive sensors simple and the read-out mechanism is easy compared to other kinds of pressure sensors, such as capacitance sensors and piezoelectricity sensors, thus they have been investigated intensively.

Here, a simple piezoresistive sensors model is built. The whole resistance (R) of the device is the sum of R_e (Resistance of the electrodes) and R_a (Resistance of the active material) as given in the following equation:

$$R = R_e + R_a$$

For a given sensor, the R_e is a constant value and the change in R_a is the main source of the electrical signal change. As the resistance (R) of bulk material (Fig. 1a) can be described as the following equation.

$R = \rho L/A$ (ρ is the resistivity of the material, L and A are length and area, respectively.)

From the equation we can conclude that the change in R_a is mainly derived from three factors: First is the geometry deformation of the bulk elastomer composite. The L and A parameters change as the deformation of the material, leading to the variation of resistance value; Second, the resistivity of semiconductor filters and conductor filters owing to changes in the semiconductor band structure and interparticle separation. In these sensors, a low compressive modulus of the sensing element favors a high sensitivity, as larger deformation can be assumed in low modulus materials at a given pressure. However, for a given material, the compressive modulus is a constant value. To improve the sensitivity, porous structures [13,14] are generally applied as modulus is lower and can be regulated in a large range. In addition, the stress is concentrated at the pillars between pores, resulting in large deformation and better response. Zhao et al. [13] reported a hollow-structured pressure sensor based on graphene–silicone-composite, in which the resistance and mechanical modulus of the composite can be tuned separately. Thus, the sensor sensitivity and linear range can be regulated, reaching a sensitivity of 15.9 kPa^{-1} in

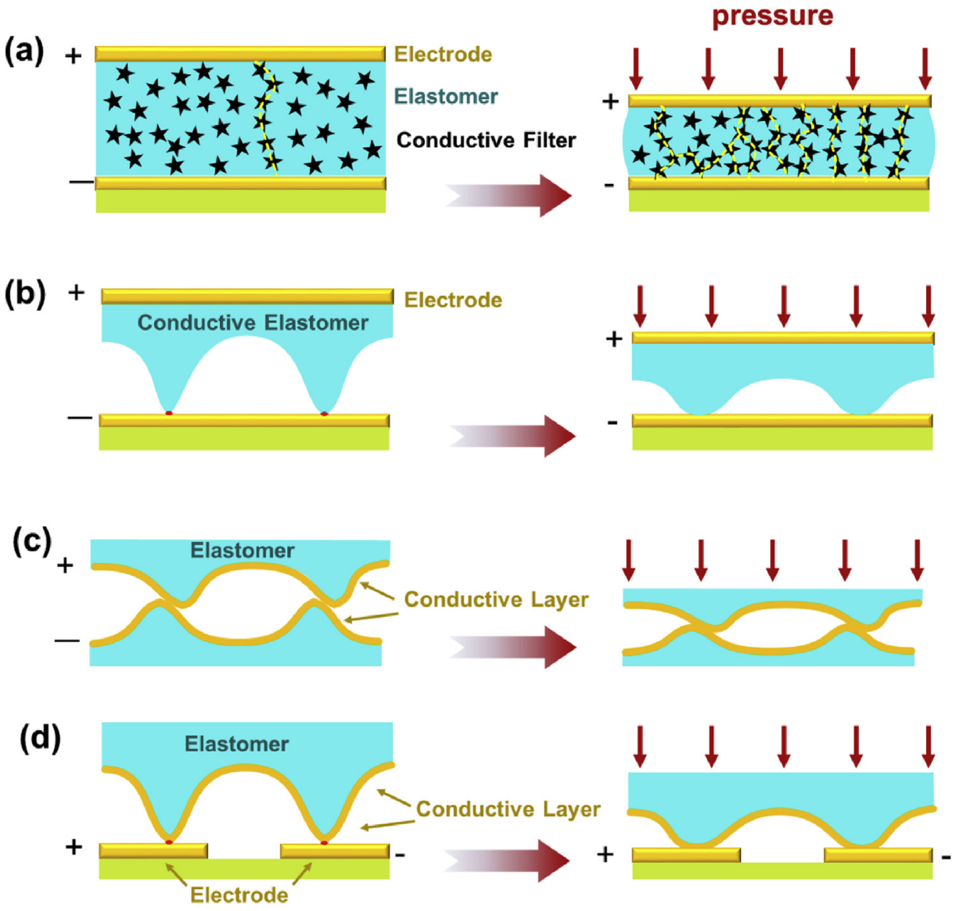

Fig. 1. The schematic images of the transduction structure of different resistance pressure sensors: (a) bulk piezoresistivity sensor. (b), (c) and (d) are contact resistance type of sensors.

0–60 kPa linear region by carefully structure tuning process. The microstructure can help the concentration of stress in a sharp region, which increases the deformation of elastomer (Fig. 1b). In addition, the large contact resistance change can further increase the sensitivity. Nowadays, microstructure is one of the most commonly used techniques to improve performance of piezoresistive pressure sensors, especially for contact resistive pressure sensors. As shown in Fig. 1c, a typic contact resistive pressure sensor is constructed by two face-to-face microstructure elastomers, which micromorphology surface is modified by conducting layer. Zhang et al. [15] presented an ultrasensitive pressure sensor based on the silk-molded PDMS-SCNT composite materials. The sensor demonstrated very low detectable pressure limit (0.6 Pa) and high sensitivity (1.80 kPa^{-1}). The advantage of this symmetrical face-to-face structure sensor is that it provides ultrahigh sensitivity at low pressures range and insensitivity to temperature and can be easily made flexible. However, despite active material, the conducting layer also acts as electrodes, the low conductivity electric properties limit its sensitivity in high-pressure range. To improve this, a new elastomer-to-electrode structure is developed (Fig. 1c). The high conductive metal electrode ($R_e \ll R_a$) efficiently transmits electrical signal change with negligible effect, which provides the sensor with high sensitivity and signal-to-noise ratio, as well as a large working range. Chen et al. [16] reported an elastomer-to-electrode structure sensor with wide linear range of pressures (from 0 kPa to 50 kPa) and unprecedented high sensitivity (1851 kPa^{-1}).

2.2. Key parameters of pressure sensors

As a transducer, the piezoresistivity sensor has to transform the imposed force into an electrical resistance signal. Several key parameters are often used to quantitative evaluate the performance of the device. These parameters include the sensitivity, sensing range and linearity, the limit of detection, response time and recovery time, operating voltage, hysteresis, and stability.

Sensitivity, defined as the slope of the resistance relative change versus applied pressure curve, is the key performance parameter in pressure sensors. A high sensitivity pressure sensor usually presents a very low detection limit and can yield a very high signal-to-noise ratio (SNR) signal, enabling the device to detect subtle pressure changes, which is very important for accurate pulse monitoring. Sensing range and linearity are also two key parameters for pressure sensors. On the one hand, a large sensing range greater than 10 kPa is required to mimic the pressure sensing performance of human skin [16,17]. On the other hand, many important practical applications, such as pulse monitoring, require the device to perform on a large dynamic range. However, most of the high sensitivity sensors exhibit a very small sensing range, to balance the sensitivity and sensing range remains great challenge. Linearity is the deviation from a straight regression line of sensing curve. Good linearity responses of pressure sensors signify a more simple data fitting. The response and recovery time are two important parameters to evaluate the real-time dynamic pressure sensing performance of pressure-sensing devices, which are important for high-frequency signal detecting. A low operating voltage can reduce the power consumption of device, thus can improve battery life of wearable system. Hysteresis is the parameter to measure the difference in the signal versus pressure curve under loading and unloading of pressure. This is an important drawback in almost all the pressure sensors, and the effect should be minimized in practical application. Stability, the parameter to evaluate the long-time sensing ability, is defined as the change of the signal during repeated loading and unloading process.

3. Materials and devices design

Sensing components, signal transfer components, and substrate are the basic components of a typical pressure sensor. To develop a pressure sensor with comprehensive excellent sensing capabilities, such as high sensitivity, wide sensing range, good stretchability and high resolution, each component of the sensor should be carefully considered. The different performances of materials are required for different components. For example, high sensitivity is needed for sensing components to transform the outer pressure stimulus to the resistance signal. Low resistivity is required for signal transfer components. Flexibility and stretchability properties are crucial to the fabrication of skin like soft devices. Therefore, developing suitable functional materials with appropriate mechanical and electrical performance is critical to high-performance pressure sensor design and fabrication. Here, we review the development of active materials, substrate materials and electrode materials for sensing components, substrate and signal transfer components in the following section.

3.1. Active materials

The active materials are key components for devices to sense the external pressure stimulate. For high-performance pressure sensors, the ideal active materials devices should possess good electronic, exceptional chemical stability, good mechanical compliance, and compatibility with large-area processing techniques. Nano conducting materials, including, polymeric semiconductors, graphene, CNT and conductor or semiconductor nanowires, are most commonly used active materials realized by embedding into or placing on the elastomeric polymer substrates.

The flexible, π-conjugated, carbon-based conducting polymers are materials that have shown great promise for soft device fabrication. Semiconducting and conducting such as polymers polypyrrole (PPY), poly(3,4-ethylenedioxythiophene) poly(styrenesulfonate) (PEDOT:PSS) [18,19], polyaniline (PANI), and their derivatives are most commonly used polymer for flexible electronics fabrication. This class of materials is compatibility with tradition solution processing techniques thus they could be processed by spray and print techniques and offer low-cost, large-area device arrays [20,21]. In addition, compared to inorganic counterparts, their mechanical properties are more comparable to soft human skin, making them ideal for implementation in skin-like device design. However, their stability and electronic performance can't yet compare with that of metal, graphene, CNT and other inorganic conductors or semiconductors.

Metal materials, such as metal nanoparticles [22], metal nanowires [23–30], metal nano-thin films [31], and liquid metal [32–37] are the most widely used materials flexible device. These materials can be used as active materials and electrodes due to their excellent conductivity. Notably, high-aspect-ratio one-dimensional (1D) nano metal materials are most widely used in stretchable devices as their ability to form and maintain their percolation network at relatively low concentrations even under large strain. A high level of conductivity can be maintained when these composites under strain, as 1D conductive fillers tend to slide against each other rather than getting fractured, thus the percolation pathways are preserved. In addition, when 1D conductive fillers hybrid with 2D or 3D nanoparticles, the resistance of the conductor under strain can be further improved. For example, when Ag nanowire and Ag or Au nanoparticle were used as composite conductive fillers (Fig. 2a), the nanowire bridged practical to maintain percolation paths under strain. As the high oxidation tendency of Ag, Ag nanowires are easily oxidized when exposure in air, which limits their long term applications [23,26,27,38]. Furthermore, highly corrosive of Ag in biological environments hinder their applications in bioelectronics. Kim. et al. [23] reported highly conductive gold-coated silver nanowires, which present good stability and biocompatible.

Carbon nanomaterials, such as CNTs [39–44], graphene [13,42,45–58], and carbon black [59], have been remarked for their exceptional chemical stability and biocompatible properties, as well as their good electronic and mechanical.

The 1D CNTs are the most widely used types of fillers for their high anisotropy property and excellent conductivity. Another advantage of CNTs is that they can deposit onto flexible or stretchable substrates directly through solution processing techniques, which is crucial for its commercial process. Spin-coating [60–62], dip-coating [63,64], spray-coating [65], vacuum filtration [15], and inkjet printing [21] are the most commonly used solution deposition techniques. Many new techniques, such as contact and roll printing, mechanical shear techniques, Langmuir–Blodgett (LB) method [66], have been developed to achieve large-scale alignment high-performance CNTs film [67,68]. Recently, Chen et al. [69] have developed a mass-producible and all-solution processable technique by combining spray and Langmuir–Blodgett (LB) method for large-area high-performance carbon nanofilm, including CNTs, carbon black and graphene film. In addition, the self-assembly technique that can align 1D nanomaterials into an interconnected network or designed micropatterns has been a key issue for its applications in device design. Currently, the most widely used 1D nanomaterials aligning technique include the methods of tri-phase contact line manipulation, Langmuir–Blodgett film technique, and solution mechanical shearing approaches.

Graphene, the single sp^2-hybridized carbon atom-thick 2D material is the world's strongest, stiffest material, and thinnest as well as being an excellent electron conductor, is another ideal alternative active material for constructing of the pressure sensor [70–76]. Depending on its processing technique and performance, graphene can be used as a conductor or semiconductor. Since recent reviews on graphene have discussed its properties and applications thoroughly, here we focus on its related properties and a few application examples of pressure sensors. Chemical exfoliation, mechanical exfoliation, and CVD techniques are the most popular processing techniques: Chemically exfoliated including the oxidation-reduction process in liquid, which leads to the mass production of low-cost graphene sheets. These products are compatible with solution-processing techniques, including vacuum filtration, spin-coating, spray coating, dip-coating and inkjet printing, which are key elements for large-scale production [47,49,55,56]. However, numerous defects would introduce during the chemical treatment process, thus leading to the reduction of electrical and optical performance. Mechanical exfoliation, including sonication and high-shear mixing technology, is another scalable method for mass-producing of defect-free graphene nanosheet [16,50,53]. CVD method can grow large-sized high-quality graphene films with excellent electronic qualities on metallic or SiC surfaces. However, when transferring such films to soft substrates, contaminants and defects may be introduced, which limits its applications [13,17,45].

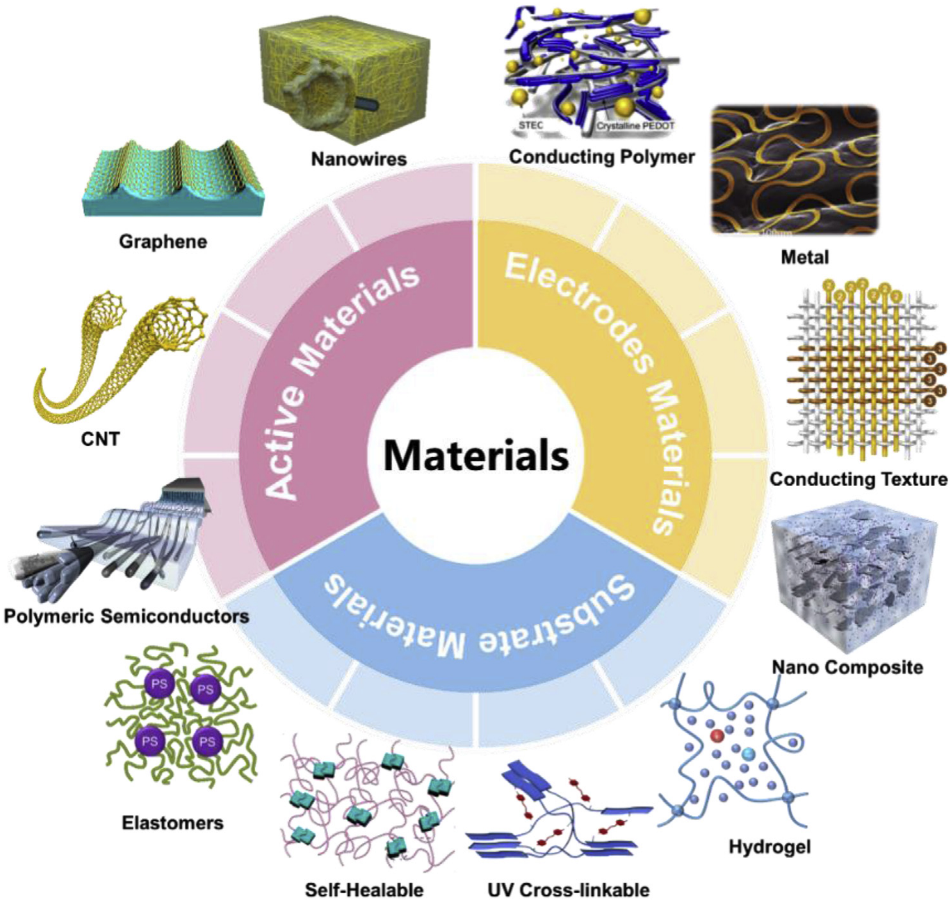

Fig. 2. Overview of materials for flexible and wearable pressure sensor devices design. These materials can be classified into three kinds of materials: Active materials could response to pressure and transducing this stimulate into an electrical signal. Images reproduced with permission: "Polymeric Semiconductors" [20], Copyright, 2019, Macmillan Publishers Ltd.; "CNT" [116], Copyright, 2019, RCS; "Graphene" [16], Copyright, 2019, Elsevier Ltd.; "Nanowires" [23]. Copyright, 2018, Macmillan Publishers Ltd.; The highly conductive electrodes materials play a key role in electrical signal transduction. Images reproduced with permission: "Conducting Polymer" [19], Copyright, 2019, AAAS; "Metal" [84], Copyright, 2013, Wiley-VCH; "Conducting Texture" [96], Copyright, 2016, Wiley-VCH; "Nano Composite" [38] Copyright, 2017, Macmillan Publishers Ltd.; The flexible and stretchable substrate material is an important factor in the development of the skin like soft pressure sensors. Images reproduced with permission: "Hydrogel" [105], Copyright, 2018, Macmillan Publishers Ltd.; "UV Cross-linkable" [21], Copyright, 2018, Macmillan Publishers Ltd.; "Self-Healable" [124], Copyright, 2018, ACS; "Elastomers" [97] Copyright, 2019, ACS.

3.2. Electrodes

Stretchable electrode materials are the foundation for the soft smart wearable devices. The application of metal materials, carbon nanomaterials, ionic hydrogel and conducting polymers have been as stretchable electrodes have been intensively investigated. The ideal stretchable electrode materials could maintain high conductivity under large strains and possess excellent stability. Among them, carbon nanomaterials have been introduced above, and the remaining stretchable electrode materials are mainly analyzed here. The hydrogel is envisioned to be implemented in applications such as smart robotics and wearable electronic devices due to its high stretchability, transparency and good biocompatibility [77–83]. Usually, a hydrogel contains a highly elastically polymer network and water. As liquid water can conduct ions, thus hydrogel can serve as a stretchable ionic conductor. Wang's [81] group reported a high transparent and stretchable soft STENG composed of ionic hydrogels and PDMS films. The STENG could be designed into arbitrary complicated shapes for energy collection and tactile sensing. Sun Jeong-Yun [79] and co-workers reported a highly stretchable ionic touch panel based on transparent based on hydrogel electrodes. The material is with 98% transmittance for visible light and it could be used on skin to write words, play the piano or playing games. However, the development of ionic conductors is limited due to its low conductivity and difficult to package, the next generation of hydrogel devices should have high-precision patterning and good conductivity for better performance.

Furthermore, metal electrode materials with the specific design are suitable for the stretchable electronics, such as Au on the PI substrate [9,84], Ag or Cu on the stretchable materials, etc. [18,23–30,32–38,85–88] Although the material itself has low stretchability, its specific structure makes the device highly stretchable. For example, Rogers' group [84] reported multifunctional epidermal electronic systems based on the thin, filamentary serpentine conductive traces with an open mesh layout. These unique designs provide large deformability at the level of the overall system with the negligible effect of its elastic moduli. Moreover, Pan's [12] group fabricated high stretchable and transparent patterned Ag nanofibers, which was fabricated by electrospinning and sputtering. The conducting fiber could be patterned by photolithography techniques. A stretchable 8 × 8 cross-type triboelectric sensor array was prepared, which could as a self-power e-skin for human-machine interaction. Meanwhile, compounding the elastomeric materials with conductive fillers, such as carbon, the polymer [18,38,89,90] and nano metal materials [26,27,34,35,37,85,88], are a good method and improves stretchability while also having excellent electrical conductivity. Bao's [19] group reported a highly stretchable PEDOT based conductor through incorporating ionic additive assisted stretchability and electrical conductivity enhances which realize exceptionally high conductivities of up to 4100 S/cm under 100% strain. Meanwhile, printable stretchable electrode materials are especially convenient for the production of large-area electronics. Someya and co-workers [38] reported a printable Ag-fluorine rubber-based conductor with high conductivity (6168–935 S/cm) and stretchable (400%).

In addition, fabric-based electrode materials have received much attention due to their good gas permeability and washing ability [24,91–96]. For example, Zheng's [96] group fabricated a direct weaving of polyimide (PI)-coated Cu-PET (PI-Cu-PET) weft yarns and Cu-coated PET warp yarns materials. It takes advantage of producing an effective triboelectric charge, high sensitivity, washing durability and ease of industrial production. Overall, various electrode materials could be developed to provide better applications in sports, healthcare sectors, and many other fields.

3.3. Substrate materials

Substrate materials are an integral part of stretchable electronics. These substrate materials are used more frequently, including the Polydimethylsiloxane (PDMS), block copolymer (BC) elastomers [97], self-healing materials [98–104], hydrogels, and UV cross-linkable materials [21,82]. Due to their outstanding chemical stability, high transparency and flexible mechanical properties, PDMS is one of the most widely used substrate material for stretchable electronic devices. The triblock copolymer BC elastomers are usually constructed by a long soft chain in the middle and two short rigid chains at the ends. Typical BC elastomers include SBS, SEBS, SIBS. Their advantages include easy to dissolve in common organic solvents, good viscoelasticity, and outstanding fluidity. Therefore, they usually meet the physical properties of stretchable electronic devices by binding most substrates (metal, glass, and polymer) and compatible with printing technologies. Self-healing substrate materials can be roughly divided into two types: external and intrinsic. Their self-healing process is caused by the dynamic balance of the cross-linking network and the osmotic pathway in the polymer system. Based on self-healing materials, some groups have designed mechanically robust elastomers for building self-healing soft electronic systems. As a highly stretchable and transparent ionic conductor material with a three-dimensional network structure, hydrogels can be active both as active materials and substrate materials in flexible sensors [105]. Ge and co-workers [78] reported a self-patterned hydrogel-based pressure sensor, which is highly stretchable transparent and good biocompatible. In this device, hydrogels act both as soft substrate materials and conductive active materials. Hydrogels are an integral part of life. The body or the tissues of many animals and plants are hydrogels, and the conducting mechanism of living matter and hydrogels are both using ions, thus hydrogels can be used as the biocompatible materials to connect metallic electrodes and living tissues. In addition, the elastic moduli of hydrogels materials are also close to living tissues and highly tunable beyond from 1 kPa to 100 kPa. Furthermore, the self-healable property of hydrogels, combining with its soft feature, endow these materials great potential in mimicking the mechanical and chemical properties of human skin. However, despite their advantages, to develop hydrogel ionotronic devices that are strong and stretchable adhesion, water retention and fatigue resistance like skin remains great challenge. UV Cross-linkable elastomer materials can compatible with advanced photolithography [77,82] and 3D printing technologies, which can amenable to large-scale and low-cost fabrication processes. Bao's group [21] developed a UV (254 nm) cross-linking agent with two perfluorophenyl azide groups, which could react with aliphatic hydrogens of SEBS. The above-mentioned substrates have shown broad application prospects in stretchable electronic devices.

3.4. Structure of piezoresistivity pressure sensor design

3D microstructures and sensing layer are two key elements of piezoresistivity pressure sensors, which determined the most important performance parameters, including sensitivity and sensing range, of pressure sensors. The most commonly used method to the improvement of the overall performance by the development of novel elastic microstructure geometries. Despite great achievements in high-performance piezoresistivity pressure sensors development through designing novel 3D microstructures, few efforts have been devoted to study and optimize the sensing layer of the sensor. Here, we summarize the recent advances in 3D microstructures and sensing layer design in subsequent sections.

3.4.1. Microstructures geometrical design

Sensitivity and sensing range are two key parameters for measuring the performance of pressure sensors. Sensitivity is defined as the slope of the electrical signal-pressure curve (electrical signal including capacitance, resistance, current, or voltage signal). A high sensitivity sensor can perform a large electrical signal even under very low applied pressure. In many application scenarios, such as artery pulse or sound monitoring, the sensor needs to output a large signal-to-noise ratio (SNR) signals when detecting very subtle pressure changes. Many previous reported high sensitivity pressure sensor can perform under low sensing range. In bulk piezoresistivity sensor, sensitivity is generally determined by the compressive modulus and conductor filler of the composite materials. As for contact resistance sensors, microstructured elastomers, such as hemisphere [106,107], pyramid [55,87,108], nanowire [31], hierarchically [17,42,109], and epidermis inspired microstructure [52,110,111], are the most commonly used techniques to improve or regulate the sensitivity and working range of devices. Comparing to unstructured elastomers, there is more free space in the elastomers and electrode for the microstructured elastomer to expand into. Furthermore, the rationally designed geometry of microstructured elastomers can concentrate the stress on a particular region, thus making the elastomers more sensitive to weak pressure and furthermore increasing its sensitivity. For example, Ko et al. designed a hemisphere structure to enhance the sensitivity of pressure sensors [106]. In a series of work, they systematically investigated the relationship between the geometry of pyramidal microstructures and the sensitivity of the sensor [55,87,108]. As shown in Fig. 3a many other microstructures are used to increase the sensitivity of the sensor. However, most of the above sensors can only maintain its high sensitivity at low-pressure range (usually at 0–1 KPa). As the applied pressure increase to several kPa, the sensitivity decreases dramatically, which largely limited its practical application. For example, as most arterial trees are deep under skin, most of plus mechanical wave is fade-out during its propagation path in soft tissue, thus leading pulse signal extremely weak on our skin surface. Gentle pressures (up to $\approx$10 kPa) application to force the sensor close to arterial to reduce the attenuation and improve signal to noise ratio one of the solutions to overcome such problem. Therefore, developing a high-sensitivity with large sensitive range (>10 kPa) sensor is required. On the other hand, human skin can sense on a large dynamic range, to mimic its tactile sensing properties, sensing range of high sensitivity device greater than 10 kPa is urgently needed.

Usually, a relatively larger size microstructure is needed to

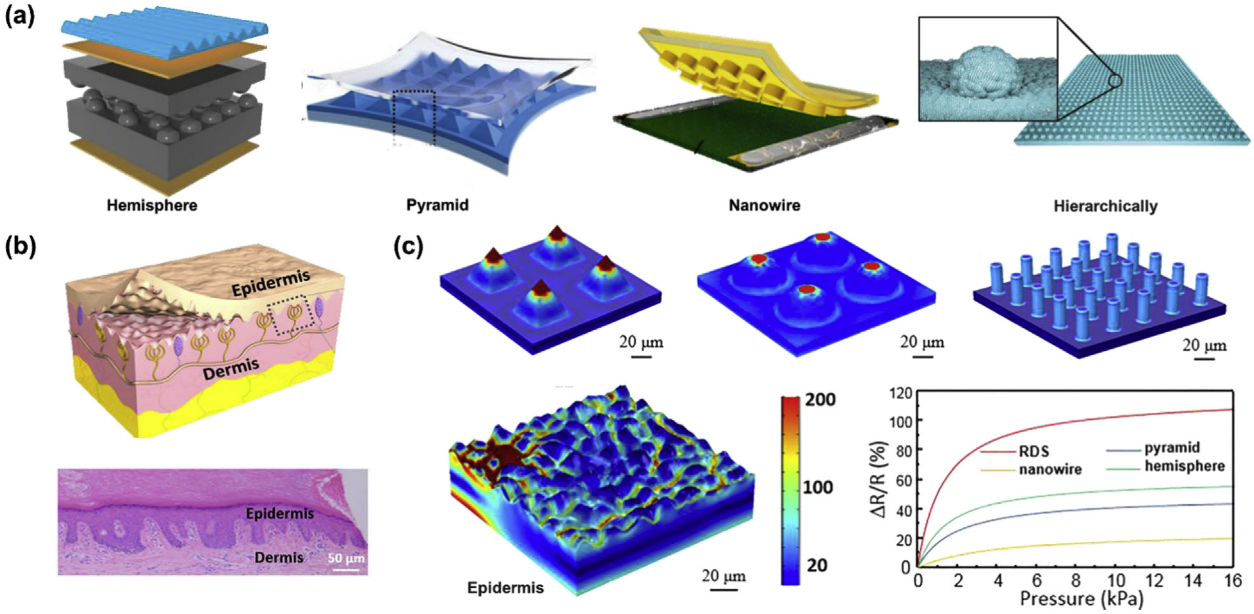

Fig. 3. (a) Various geometrical structure for high-performance pressure sensor design. Images reproduced with permission: hemisphere [125], Copyright 2015, AAAS; pyramid [108], Copyright, 2014, Wiley-VCH; nanowire [31], Copyright, 2015, ACS; hierarchically [17], Copyright, 2016, Wiley-VCH; and epidermis inspired microstructure [110] (b) Copyright, 2018, ACS; (c) The simulation results of resistance variation versus applied pressure for the above geometries. The results demonstrate the epidermis inspired microstructure perform the highest sensitivity.

increase the deformation space of the elastomer thus to enhance sensing range. However, high sensitivity property needs the microstructure elastomer as sharp and small as possible thus it could respond to subtle pressure. the inherent contradiction makes it a great challenge for the sensor to maintain high sensitivity in a wide linear range.

In order to balance the trade-off between linearity region and sensitivity, Cho and coworkers fabricated a piezoresistive sensor based on hierarchical microstructure array [17]. This unique hierarchical microstructure structure is just like the lotus leaf microstructure, featuring small protuberances on large hemispherical domes surface (Fig. 3a). Comparing to conventional domes with a smooth surface. this hierarchical microstructure performs a high sensitivity (8.5 kPa^{-1}) on a wide linear working range (0–12 kPa). As is well-known, human skin is highly sensitive to pressure over wide sensing range, inspired by the epidermis microstructure on the skin, Ren et al. [52,110,111] constructed a pressure sensor by two interlocking epidermis like microstructure PDMS layers. Computer simulation results show that the spinosum microstructure contributes to the high sensitivity (25.1 kPa^{-1}) and random distribution of increase the linearity sensing range (0–2.5 kPa). They mechanism analyses demonstrated the epidermis microstructure has advantages in balancing the trade-off between sensing region and sensitivity.

3.4.2. *Sensing layer design*

Instead of developing novel microstructures geometrical features to the improvement of the performance of the sensor, Chen and Pan's group [11,16] point out that the sensitivity and linear range can also be regulated by optimizing the conductivity and physical thickness of the sensing layer on microstructures. An idea conduct layer should with low resistance, nanoscale thin structure and mechanical durable that can easily self-aligned to the elaborate 3D microstructures. Simulation and mechanism analysis indicates that the high conductivity and thinner sensing layer contribute to the high sensitivity and large linearity: On the one hand, high conductivity sensing layer reduced signal attenuation, which increases the signal-noise ratio and sensitivity; On the other hand, the relatively thin sensing layer could increase deformation of the macrostructure, which enlarges the sensing range. To construct a thin and high conductivity sensing layer on microstructures elastomer they developed self-assembled and self-attached graphene. The sensor achieved an unprecedented high sensitivity (1473.5 kPa^{-1}) in large linear sensing range (0–20 kPa). (Fig. 4a and Fig. 4b).

This advanced sensor is advantageous in weak arterial pulse signal monitoring under wide applied pressures range from 1 kPa to 10 kPa. Notably, from the frequency spectrum of the highly accurate pulse wave signal, they observed new weak signals not reported early, which may provide valuable diagnostic information (Fig. 4c and d). In addition, the wide sensing range feature enables the device to detect high accurate pulse signals in practical scenarios, even during cycling, walking and running (Fig. 4e).

4. Challenges and outlook

4.1. *The rise of E-textile pressure sensor and its challenges*

Textile, the artificial outer skin of a human, has been widely used for thousands of years. Nowadays, textile has attracted broad interests in wearable electronic fields as its advanced breathable, comfortable and biocompatible features. Thus, piezoresistive electronic textile (E-textile) has attracted broad interests in wearable electronic fields [24,91,93]. These fiber assembly materials can dissipate the stress by fiber sliding instead of strain, therefore able to accommodate severe or complex deformation, such as bending, twist and stretching. More importantly, as for the pressure-sensing device, the hierarchical microstructure feature of the textile could significantly increase its sensitivity and sensing range, thus the textile material is an ideal candidate for constructing of the high-performance pressure sensor. For example, Cheng et al. [24] constructed a sandwich structure flexible pressure sensor based on ultrathin gold nanowire-impregnated tissue paper and PDMS elastomer films. This device presents an ultrahigh sensitivity

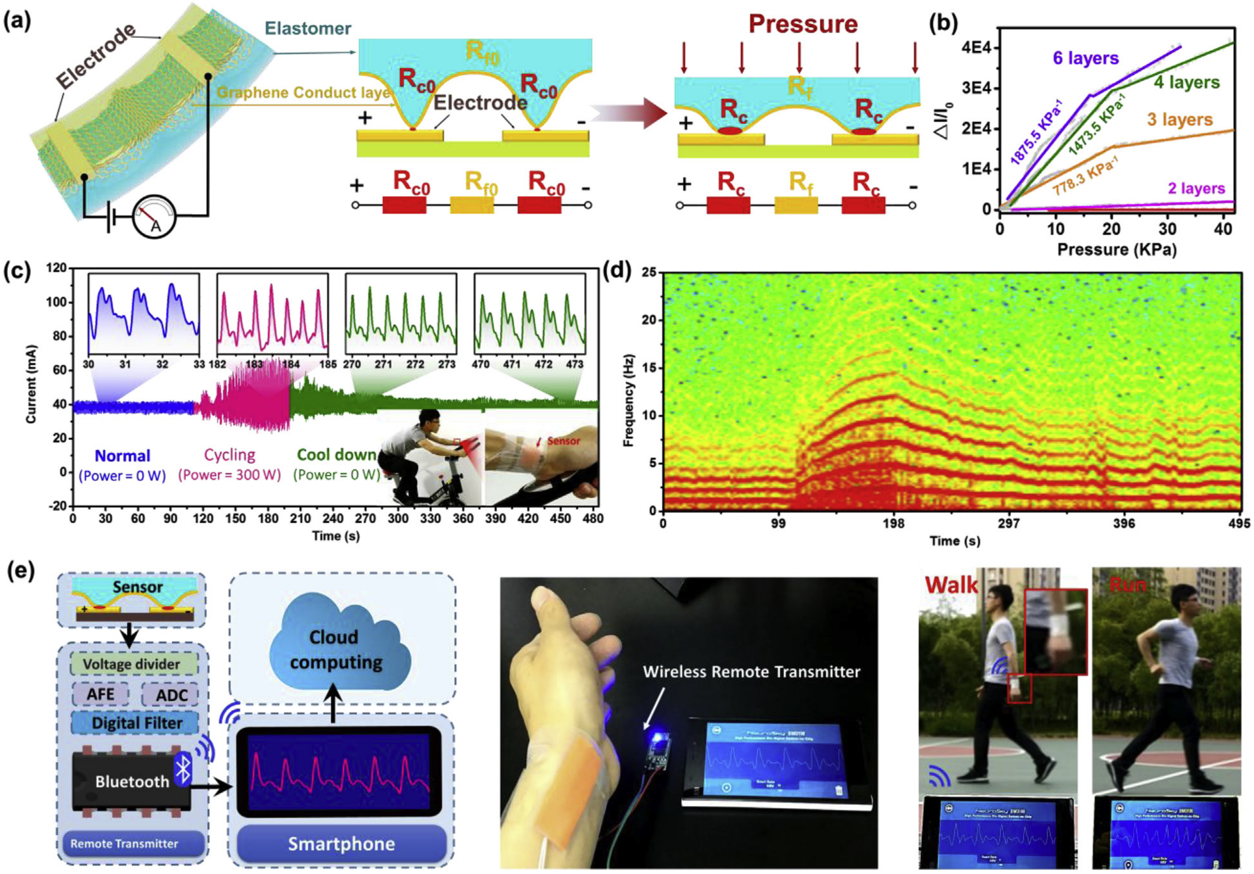

Fig. 4. Graphene-based ultrahigh sensitivity and large linearity pressure sensor: (a) Schematic of the pressure sensor; (b) The sensitivities and sensing range of the sensor can be regulated by the graphene sensing layers. (c) The real-time artery pulse signal and its frequency spectrum (d) measured by the pressure sensor during cycling. (e) The wireless wearable health monitoring system based on the high-performance pressure sensor, which can be applied during the process of walking and running in daily life. Reproduced with permission [16]. Copyright 2019, Elsevier Ltd.

(41.14 kPa $^{-1}$) and very low detection limit (13 Pa) with fast response time (17 ms) (Fig. 5a). Wang et al. [93] fabricated large-area and carbon nanotube (CNT) and Ni-coated textile-based pressure sensors (Fig. 5b). The textile sensor achieves high fast response (≈24 ms), low detection limit (2 Pa), high sensitivity (14.4 kPa^{-1}) and wide linearity sensing range. This device presents great potential in recognize joint movement and real-time pulse waves. In order to further improve the sensitivity and sensing range, Kim et al. [94] reported multi-layered, all-textile-based tactile sensor (Fig. 5c). Despite the textile's hierarchical structure provide more contact area and point between textile under pressure. The rational designed multi-layered structure can further facilitate a significant increase and distribute stress, which endows the sensor with improved sensitivity (26.13 kPa^{-1}) and larger linearity pressure range (0.2–982 kPa).

Benefited from the breathable, comfortable and biocompatible features, electronic textile (E-textile) has attracted broad interests in wearable electronics fields in recent years [112–115]. Nevertheless, as an on-skin device, the E-textile is inevitable to survive sweating, oil, and stains from skin and ambient environment, as well as washing cycles. Thus, for the interest of fundamental research and practical applications, high-performance E-Textile that durably and fully repels stains and liquids need to be developed. Zhang and coworkers [116] designed a super-anti-fouling e-textile via coating "steel-concrete" like nanocomposite structure sensing layer on textile (Fig. 5d), which is mechanical and chemical robustness and super-repellent to a range of liquids and stains, including water, solvents, sweat, acids or even polymer paint. The as-prepared device is super-repellent to oil, water, sweat, acids, and even polymer paint, thus the device can survive under such harsh environment (Fig. 5e). Further durability tests proved that the device can withstand repeated standard machine washing cycles without visible structure damage and performance sacrifice (Fig. 5f).

4.2. Mimicking the complex human pressure sensing elements

Presently, the fabrication of high-density, high-sensitive and multi-functional detection of electronic skin is a research hotspot in which sensors play an important role. Biological skin can recognize the magnitude and direction of different stresses, and there are many studies on the stress sensors which can detect a wide range of stress, but there are few reports on the detection of 3-dimensional stress to the differentiation of normal and tangential stresses [107,117]. Bergbreiter and co-workers demonstrate a taxel geometry to create a 3-axis tactile sensor that can detect applied pressure based on the contact resistance approach (Fig. 6) [117]. As stresses are applied, the "pad" and "pillar" come into physical contact. When normal stress applied, the pillar and pads electrode expanded under pressure owing to the Poisson effect. The two sides will come into contact resulting in reduced contact resistance. However, asymmetry of contact resistance between the two sides when applying shear stress on the sensor, contact resistance along the stress direction will decline, whereas increase on the other side.

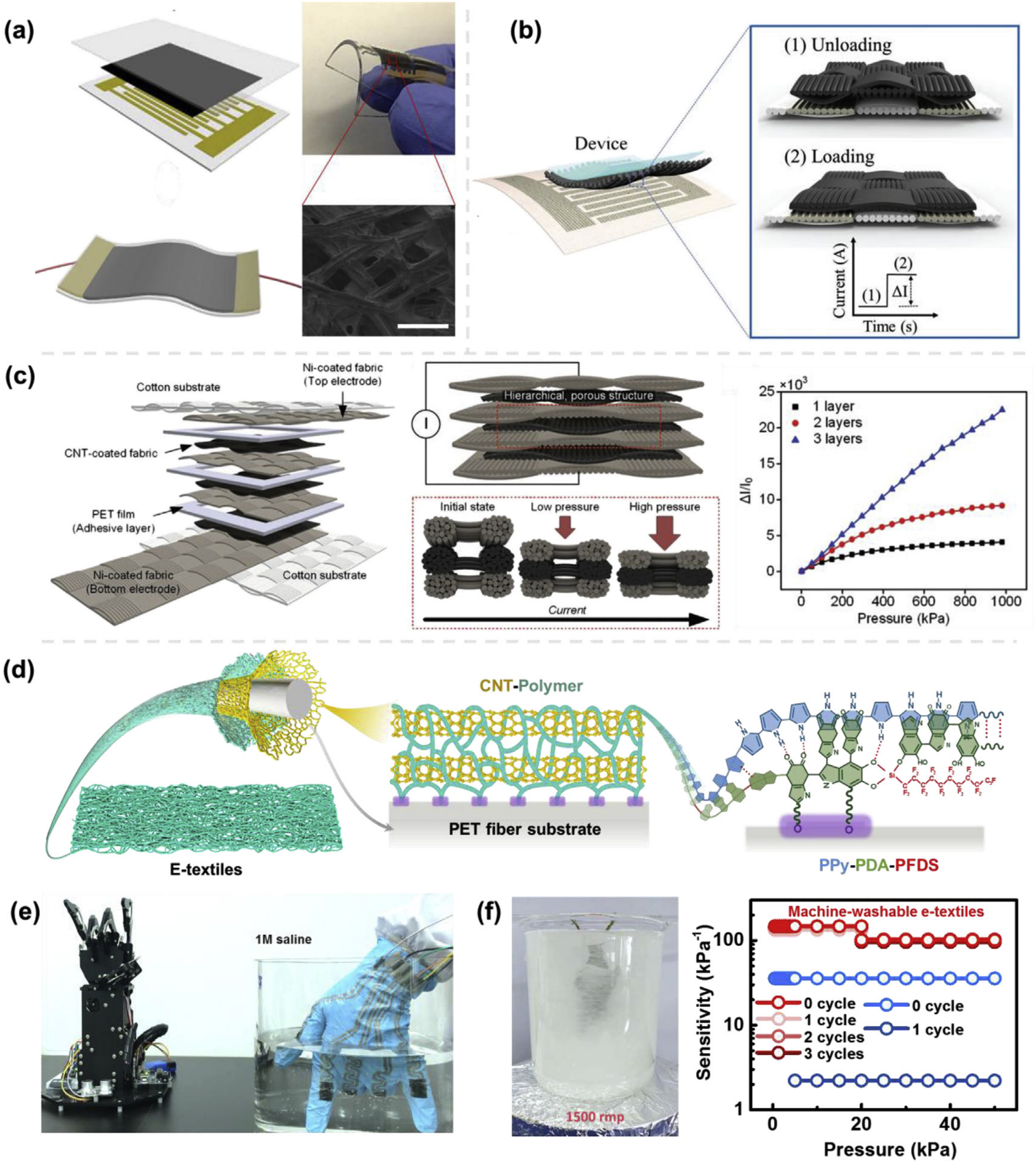

Fig. 5. Textile based pressure sensor: (a) A flexible and highly sensitive pressure sensor based on ultrathin gold nanowires coated tissue paper. Reproduced with permission [24]. Copyright 2014, Macmillan Publishers Ltd. (b) All-textile-based flexible pressure-sensor. Reproduced with permission [93]. Copyright 2017, Wiley-VCH. (c) A multi-layered structure tactile pressure sensors, which perform high sensitivity and good linearity in the ultrawide sensing range. Reproduced with permission [94]. Copyright 2019, Wiley-VCH. Liquid-repellent and machine-washable smart e-textiles: (d) Schematic illustration of the structure of the e-textiles. (e) The demonstrating of liquid-repellent property of e-textiles sensors. (f) The sensitivity of e-textiles sensors after each standard machine washing cycle. Reproduced with permission [116]. Copyright, 2019, RCS;

In order to explore its effect in practical applications, a robot of the same size as the palm of an adult human was fabricated and attached 12 taxel sensors with a total of 41 electrical leads. The robot hand presents a good shear force detecting ability in experimental tests.

Human skin can detect a variety of different physiological signals due to its various sensory receptors (including pressure, strain, PH, vibration, and temperature, etc.) [9,50]. It can still monitor the above signals and transmitting nerve impulses to the brain even in the deformation conditions. In order to mimic these properties, it's necessary to integrate kinds of sensors on the substrate to detect the corresponding stimuli and make the electronic skin possess the excellent ability to resolve various signals with deformation. For instance, Someya and co-workers [50] fabricated ultrathin, bending-insensitive and optically transparent pressure sensors in the bent state for different normal forces which were based on the uniform dispersion of conducting nanomaterials (CNTs and graphene) inside the electrospun nanofibers (Fig. 7a). The thin substrate and entangled nanostructure facility the device remain normal operation even in the extreme bending process. Moreover, real-time pressure distribution under complex deformation was demonstrated via a 12 × 12 ultrathin pressure-sensor matrix (including the substrate, nanofiber sensor, active sensor, and encapsulation). The results reveal that the sensor could provide accurate measurement of pressure without suffering from the inaccuracy induced by mechanical deformation. Another approach to

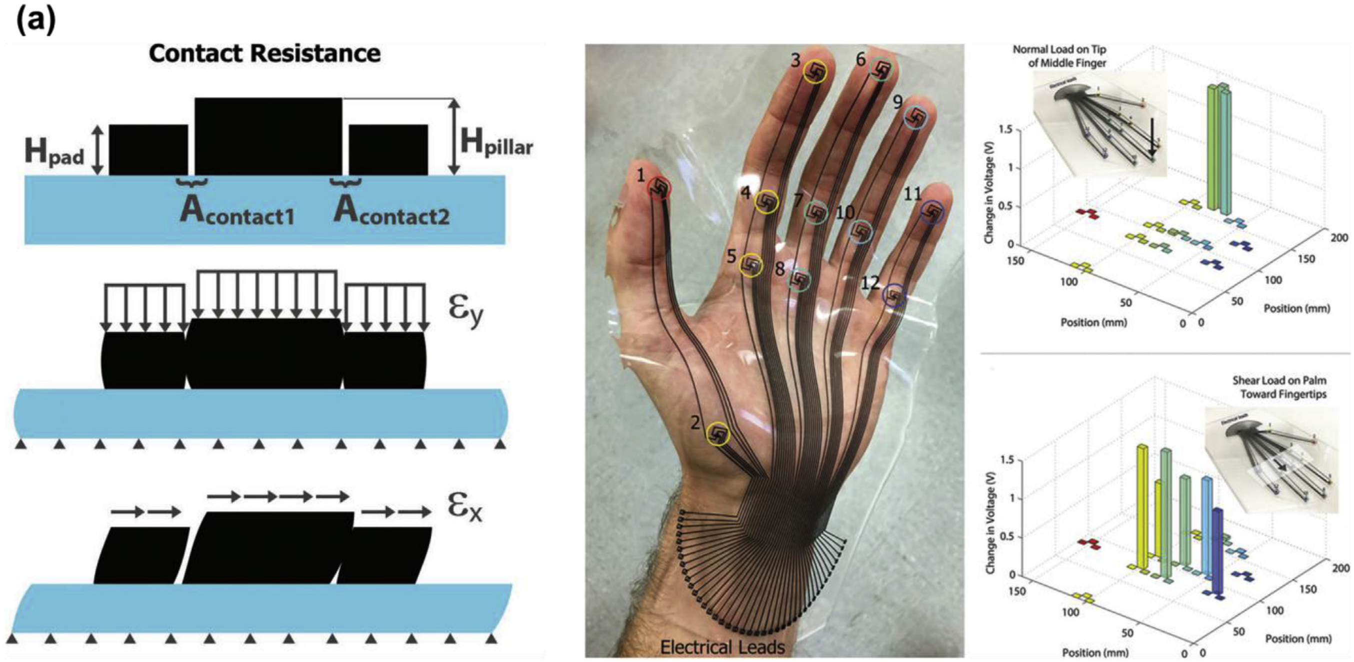

Fig. 6. Tangential force sensor with a rationally designed structure with three conductive blocks. The contact resistance between the small blocks and the center large block can measure tangential force value. The force direction can be surmisable as the contact resistance between the center large block surrounding and small blocks decrease in the direction of shear and increases on the opposite side. Reproduced with permission [117]. Copyright 2017, Wiley-VCH.

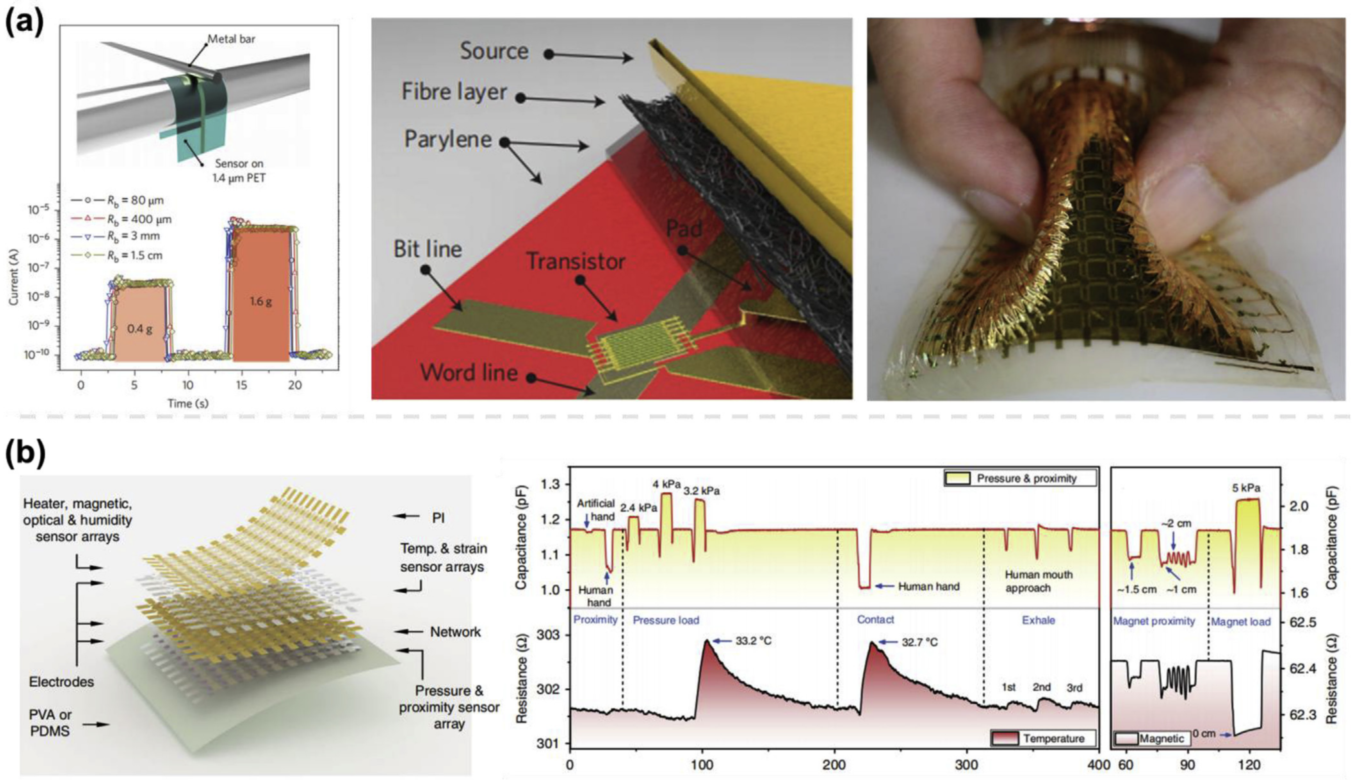

Fig. 7. (a) A flexible and extraordinarily small bending-sensitive, resistive type of pressure sensors based on ultra-thin nanofibres networks. Reproduced with permission. Reproduced with permission [50]. Copyright 2016, Macmillan Publishers Ltd. (b) Simultaneous multiple-stimuli sensing e-skin based on skin-inspired highly stretchable matrix networks. Reproduced with permission [9]. Copyright 2018, Macmillan Publishers Ltd.

integrating sensors and decoupling various signals is to fabricate the island – bridge structure. Pan's group [9] illustrates a skin-inspired multilayered highly stretchable and conformable matrix network that integrates six different types of sensor units (Fig. 7b). Such designs make the matrix capable of detecting parameters, including but not limited to pressure, strain, temperature, RH, UV light and magnetic field stimuli, from the environment simultaneously without affecting each other.

Human fingertip skin can detect small objects due to the high density of mechanoreceptors. Therefore, high-resolution sensor arrays are an integral part of emulating human tactile sensation. Additionally, high-resolution sensor arrays could be promising in fields such as intelligent robotics, prosthetics, and medical applications. Pressure sensor arrays have been reported in many previous studies. However, limited by the integration and fabrication techniques, the fabrication of large areas of high spatial resolution sensors remains a great challenge. Until now, the ultrahigh-resolution pressure sensor arrays are based on the piezoelectricity transduction method and transistor array [10,118–122].

For instance, based on vertical piezoelectricity zinc oxide nanowires, Wu et al. [123] reported addressable two-terminal transistor arrays which can convert forces into electronic signals through piezoelectricity (Fig. 8a). Strain-induced piezo-charges in the NW at the contacts can modulate Schottky barrier heights and hence change carrier transportation, which can be used as a gate signal. Consequently, the device structure is considerably simplified. The two-terminal devices could attain a high 35 times higher density than human fingertip skin. In addition, the devices were observed to respond to strain ranging from a few kPa to 30 kPa. Pan et al. [10] reported a pressure sensor array based on nanowire light-emitting diode (Fig. 8b). Each pixel is composed of a single n-ZnO nanowire/p-GaN light-emitting diode, the light intensity of which can be tuned by strain through piezo-phototronic effect. The strain applied to devices distorts the basic unit cell of ZnO, thus inducing piezoelectric polarization charges in the junction region. The energy band is distorted by induced charges at the junction region which tends to temporarily trap holes. Hence, the carrier injection and recombination rates are increased. The light intensity of pixels under compression is dramatically increased and the pixels without compression show no obvious change when compression is applied to the device through a convex mold. In addition, the optical signals from all pixels are read out by CCD in parallel with a time resolution of 90 ms. Furthermore, the device attains a pixel density of 6350 dpi and spatial resolution of 2.7 μm. Wang et al. [21] reported a high yield and uniformity fabrication process and an intrinsically stretchable transistor array with high density through that method (Fig. 8c). The patterning process comprises two key strategies that are using a fluorinated polymer as the sacrificial layer and inkjet printing. In addition, the fabrication process achieves high overall yield of 94.4% for 108-transistor array. The

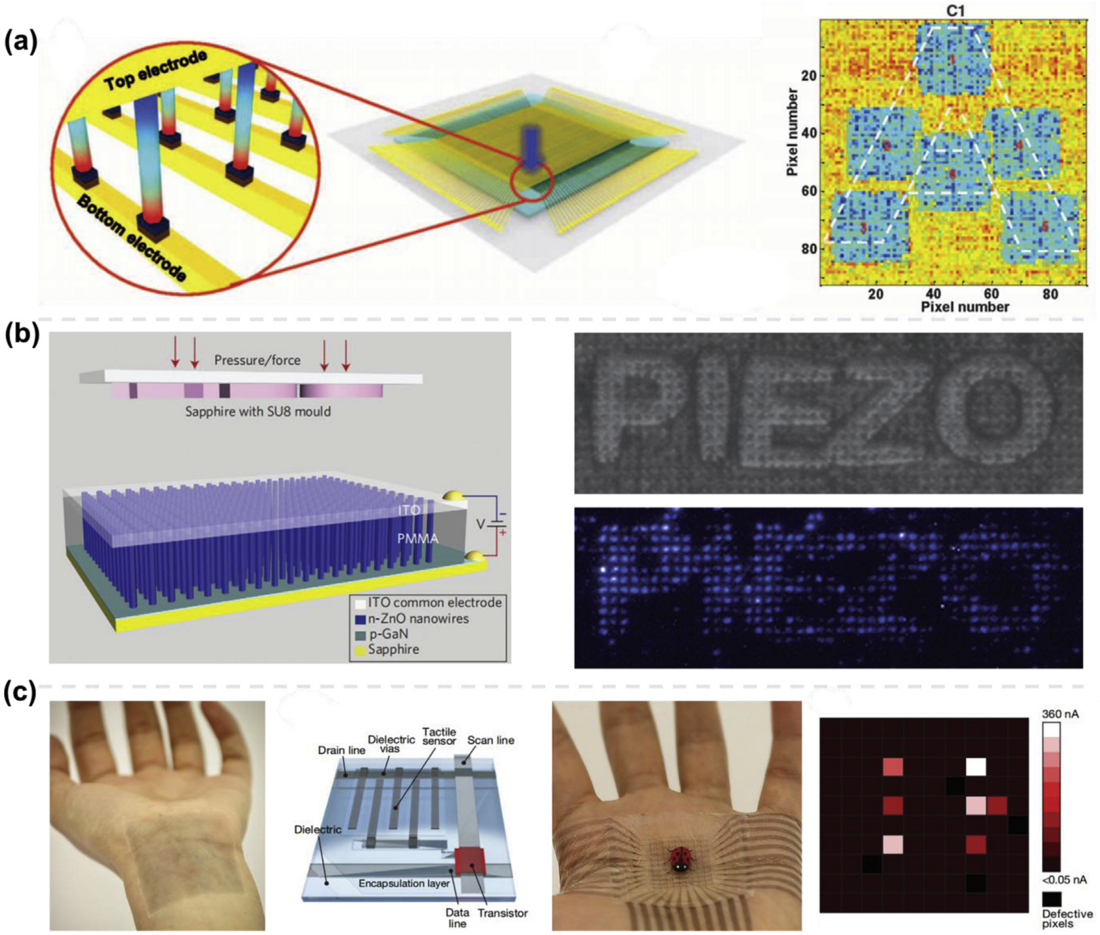

Fig. 8. High-resolution pressure sensor arrays: (a) Self-powered large-array three-dimensional circuitry integration of vertical-nanowire piezotronic transistors for high-resolution imaging. Reproduced with permission [123]. Copyright 2013, AAAS. (b) Unprecedented 6350 dpi resolution imaging of two-dimensional pressure distribution based on piezoelectric nanowire LED array. Reproduced with permission [10]. Copyright 2013, Macmillan Publishers Ltd. (c) A skin-like intrinsically stretchable high-density sensor arrays, which can accurate sensing of the position of a synthetic ladybug with six legs. Reproduced with permission [21]. Copyright 2018, Macmillan Publishers Ltd.

transistor array shows an on/off current ratio of 104 and average mobility of 0.821 ± 0.105 $cm^2 V^{-1} s^{-1}$. Furthermore, the transistor can be stretched to 100% both parallel and perpendicular to the direction of carrier transport without electrical performance decreasing. The device can attain a density of 347 transistors per square centimeter. However, sensitivity or the working range of the above high-resolution sensor arrays can't compare with the performance of human skin. Devices integrated with high-resolution, high sensitivity and large working rang piezoresistivity sensor arrays have not reported yet (see Fig. 8).

Despite the above advances, how to understand the complex tactile sense of human skin and enable the machine to mimic this feeling remains a great challenge. For example, humans hand can not only feel and weigh objects but also infer their shapes and material properties while and grasping. The diverse and complex mechanoreceptor networks, which provide sensory feedback, in the skin remain difficult for robots to replicate. Recently, Matusik et al. [95] reported a scalable tactile glove covering the full hand with 548 pressure sensors, which could measure pressure forces from 30 mN to 0.5 N. (see Fig. 9) Combining the tactile glove and deep convolutional neural networks, they built a human grasp signatures machine learning system. On the smart glove, sensors are uniformly distributed over the full hand. These sensing platforms produce large-scale data while grasping objects. By correlation the date with objects the machine can identify individual objects, including estimate weight and explore the typical tactile patterns. In addition, the health data, including plus, respiration rate, heart rate and blood pressure vital signs, also need to be systematically studied combining with big data and AI technologies.

5. Summary and perspectives

In summary, we discussed the development of wearable and flexible piezoresistivity pressure sensor devices in recent years. These soft wearable devices hold great potential applications in various fields, including prosthetics, robotics, and real-time health monitoring. For example, it can endow the prosthetics and robotics with human skin-like tactile sense, enabling the machine more powerful and humanization. Various health risks, such as cardiovascular disease, diabetes mellitus, and arthropathy, are simultaneously on the rise as the life expectancy and arriving of the aging

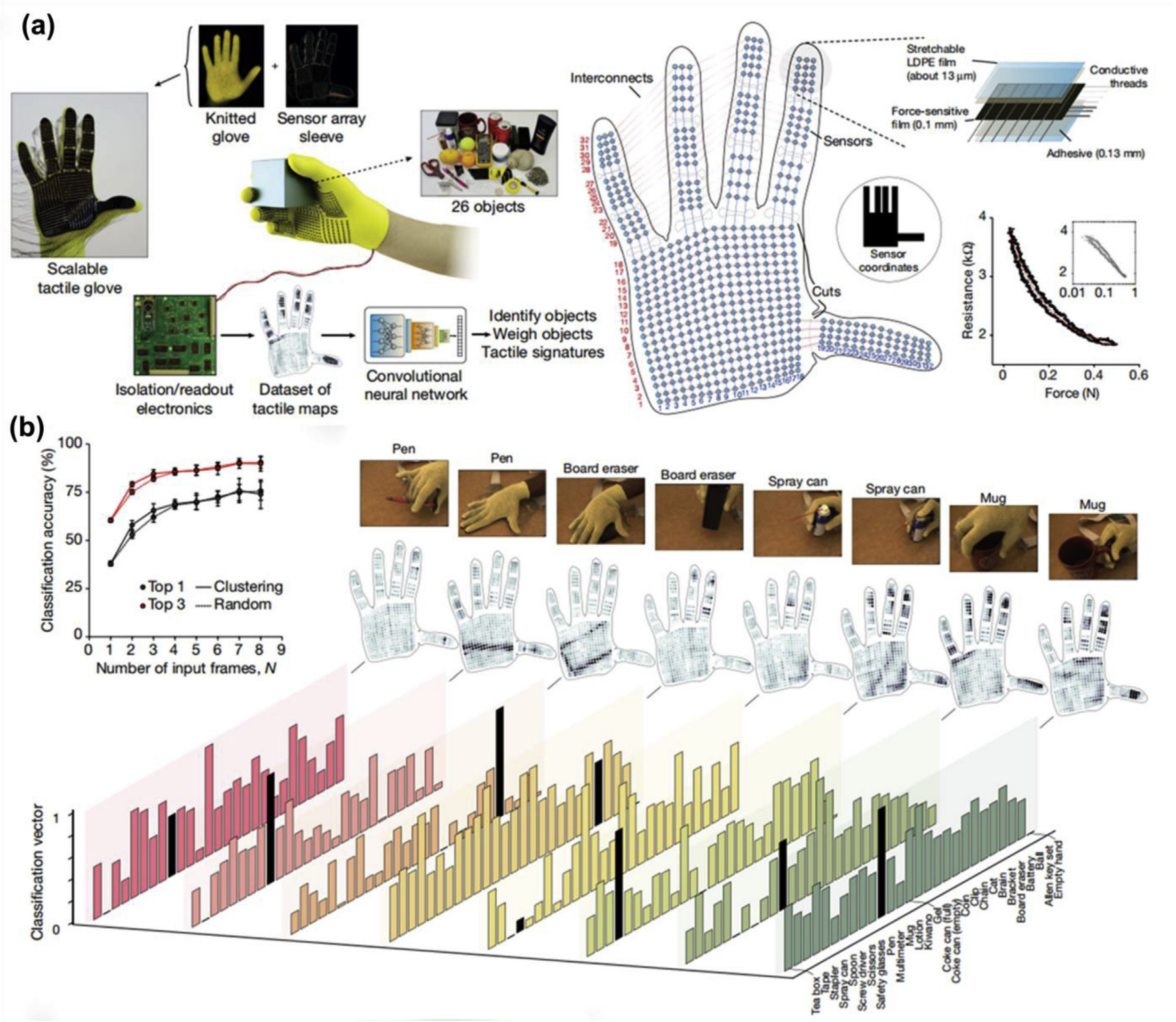

Fig. 9. Scalable pressure sensor glove for human grasp signatures machine learning: (a) Scalable tactile glove platform to learn from the human grasp. (b) The system can identify different objects accurately by learning grasp signatures. Reproduced with permission [95]. Copyright 2019, Macmillan Publishers Ltd.

population. The soft sensor can comfortably attach to our skin and continuous monitoring and improve people's health. Toward this vision, a large number of research groups over the world have been working intensely for decades, and significant progress has been achieved.

Inspired by the remarkable achievement, we summarized the various structures of the piezoresistive pressure sensor and try to provide a better understanding of the development of structure and fundamental working mechanism of the piezoresistivity pressure sensor. In this way, it may help to guide the exploration of novel structure sensors and construction of the sensors with overall high performance. Flexible pressure sensors with low detect limit, fast response speed, high sensitivity, high density, and large sensing range is developing. However, despite the progress described above, a novel and better understanding of mechanism should be explored to the development of advanced performance pressure sensor respect to overall performance parameters remains a great challenge. In addition, sensor literature specifically addressing power consumption, hysteresis, slip and force vectors has been rarely reported. The novel mechanism and structure should be further developed for slip and force vectors sensors. The fundamental origin of hysteresis has not yet fully studied and mitigation methods should be explored.

Various materials, including active materials, electrodes materials, and substrate materials, with flexibility and intrinsic stretchability for sensor constructing, have been developed. Self-healable and stretchable insulators, conductors, semiconductors and their composite that with good interfacial compatibility and processability should be further developed. To develop a skin-like multifunctional integrated system, various electronic components and high-density sensors (including pressure sensors) should be integrated into the tiny soft substrate, thus large-area and low-cost integration and fabrication techniques are required. Furthermore, a mass of information would be generated by these sensors, how to collect, organize, manage process, maintain and understand those huge signals need to be further explored.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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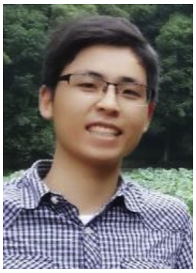

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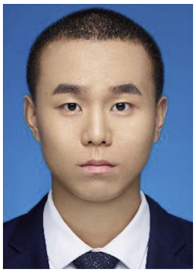

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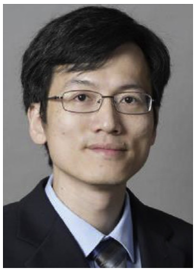

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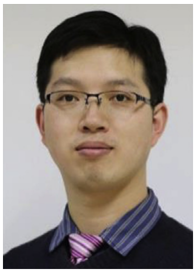

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