Recent progress in tactile sensors and their applications in intelligent systems

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Abstract

With the rapid development of intelligent technology, tactile sensors as sensing devices constitute the core foundation of intelligent systems. Biological organs that can sense various stimuli play vital roles in the interaction between human beings and the external environment. Inspired by this fact, research on skin-like tactile sensors with multifunctionality and high performance has attracted extensive attention. An overview of the development of high-performance tactile sensors applied in intelligent systems is systematically presented. First, the development of tactile sensors endowed with stretchability, self-healing, biodegradability, high resolution and self-powered capability is discussed. Then, for intelligent systems, tactile sensors with excellent application prospects in many fields, such as wearable devices, medical treatment, artificial limbs and robotics, are presented. Finally, the future prospects of tactile sensors for intelligent systems are discussed.

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1. Introduction

The development and application of intelligent systems have significantly improved the quality of people’s lives and provided great convenience in many aspects [1]. For example, recent wearable intelligent systems can collect large amounts of data at any time to determine illness [2]. At the same time, intelligent systems for human–computer interaction have been widely used in robots, electronic tablets, biometric devices, etc. [3–7]. However, intelligent systems that work in complex environments require precise coordination of various sensors. Among them, tactile sensors are the most important and complex sensors because they can be used to construct a large-area fast-response high-resolution system [8–10]. As shown in Fig. 1, tactile sensors have been already used in every aspect of daily life, and their high performance has been well studied by researchers, such as high-resolution, stretchability, self-healing, self-powered capability and biodegradability. Among them, high resolution promotes the development of human–computer interaction interface and robotics. Stretchability makes devices more similar to human skin and extends the range of application of tactile sensors. Self-healing greatly increases the service life of the devices. Self-powered capability realizes the collection of natural energy, and biodegradability provides a development direction for transient electronics. Intelligent systems made up of these tactile sensors are gradually changing the way people live and work, and with the progress of science and technology, people’s ideas will change dramatically. The tactile organs of most animals are all over their body [24] and exhibit various sensing capabilities, such as for temperature, humidity, mechanical stimulation and roughness [25–28]. Faced with the application requirements of wearable electronic devices and devices used in the biomedical field, human–computer interaction, intelligent robotics and other fields, tactile sensors are needed not only to mimic the tactile organs of human skin but also to express sensations in a quantitative way [29]. Therefore, realizing this functionality has been one of the important challenges and bottlenecks in the field of sensors for a long time [30].

Broadly speaking, for application in intelligent systems, tactile sensors possessing high performance refer to flexible electronic devices, which are generally made of lightweight, stretchable and elastic materials [31]. According to the working mechanism, tactile sensors are mainly based on the piezoelectric, capacitive, and
piezoresistive effects. Among them, the piezoelectric effect refers to the electric polarization of a piezoelectric material caused by a mechanical force [32]. Tactile sensors based on the piezoelectric effect have the advantages of high natural frequency, high sensitivity, high signal-to-noise ratio and good stability [33]. As for capacitive tactile sensors, whose operating principle is that the variation in the capacitance depends on the change in the relative position between the plates under an external pressure, they can be integrated over a large area and used at low power consumption [34]. The piezoresistive tactile sensors based on the piezoresistive effect, in which the conductivity varies under an external pressure, have the advantages of large detection, simple signal processing, and strong anti-interference capability compared with other types of sensors [35]. In addition to the three types of tactile sensors...
mentioned above, photoelectric, photoelectromagnetic, and magnetoresistive sensors are also applied in many fields for intelligent systems [36].

In the early days of the development of tactile sensors, studies about touch were limited to whether the sensor was in contact with the object or the degree of contact [36]. Followed by great efforts, more detailed works on the design, principles and methods of sensors were conducted to improve their performance [37]. To date, tactile sensors with high performance have been an essential element in intelligent systems. In the operation process of an intelligent system, tactile sensors are used to transform the physical, chemical and biological information acquired by direct sensing perception into digital information, which can be easily recognized and transmitted to the back-end platform for processing and analysis. As a typical example, Takashima and co-workers [25,38] designed a piezoelectric three-dimensional force tactile sensor, which was installed on the end of the smart finger of a robot. Marcus Meyer of Germany developed an octopus-like underwater robot with a new tactile system that accurately sensed obstacle conditions and automatically performed surveys of the seabed environment [39]. The new hairy electronic skin developed by Seoul National University in Korea can be widely used in prosthetics, heart rate monitors and robots [40].

In this review, we first focus on the development of tactile sensors with high performance, mainly high resolution, stretchability, self-healing, self-powered capability and biodegradability. Then, great potential applications in intelligent systems based on the tactile sensors with high performance are presented, such as wearable electronic devices, devices in the biomedical field, human–computer interaction systems and intelligent robots. Finally, we summarize the breakthroughs made in recent years in tactile sensors and outline the future development of tactile sensors for intelligent systems.

2. High-performance sensors

The high performance of tactile sensors has driven their development in intelligent systems. Many efforts have been made to develop new tactile sensor functionalities to satisfy different applications in intelligent systems. Material selection and structure design play important roles in achieving good performance and various functions [41]. In this section, we mainly introduce several tactile sensors based on new materials and structures that achieved high performance in intelligent systems. First, high resolution is one of the prime performance parameters of tactile sensors. The introduction of the piezotronic and piezo-phototronic effects ensures tactile sensors with high resolution. For further adaption to intelligent systems, on the basis of these two effects, new materials have been utilized to enable flexible tactile sensors under the premise of a high resolution. Second, to mimic human skin, stretchability is an essential feature for tactile sensors to realize the personification of intelligent systems. Therefore, intrinsically stretchable materials and stretchable structures are used to provide tactile sensors with stretchability. Third, tactile sensors will inevitably encounter mechanical injury in ordinary operation. Self-healing is an important function when the devices are damaged and cannot work properly. The combination of covalent adaptive bonds and supramolecular noncovalent bonds makes devices self-healing. Fourth, a self-powered capability means that tactile sensors no longer have to rely on external batteries, which enables devices with not only decreased size but also reduced natural energy consumption. Piezoelectric and triboelectric nanogenerators used as tactile sensors provide the foundation for self-powered devices. Finally, with the increasing awareness of environmental protection and the increasing needs of medicine, the biodegradability of tactile sensors plays a significant role in intelligent systems. Specific inorganic materials can endow devices with the ability to self-degrade in the environment, but some features limit their development in intelligent systems. To meet more application scenarios, breakthroughs have been made in the field of organic materials.

2.1. High resolution

Electronic devices with high resolution facilitate the development of robotics and human–machine interfaces. A high resolution enables accurate identification of the position and direction of objects by robots so that they can flexibly complete the task [42]. Additionally, tactile sensors with high resolution can effectively identify the appearance of complex objects in low visibility environments [43]. Therefore, this capability is necessary for tactile sensors to be used in electronic skin, imaging systems, intelligent signature systems and other applications.

However, ordinary piezoresistive or capacitive pressure sensors still have limited resolution of millimeter magnitude. Encouragingly, the piezotronic effect was used to improve resolution. The piezotronic effect arises as a result of the polarization of nonmobile ions in the crystal. It is an interface effect that asymmetrically modulates local contacts at different terminals of the device because of the polarity of the piezoelectric potential. The principle of strain-gated piezoelectric transistor is different from that of conventional field-effect transistors. It is realized on the metal–semiconductor contact interface through strain-induced ion polarization charge modulation local contact characteristics and carrier transport, so the piezotronic effect can improve the resolution of tactile sensors greatly. On the basis of the piezotronic effect, Wang group [44] prepared a taxel-addressable pressure/force sensor composed of a plurality of vertical ZnO nanowires between upper and lower electrodes. A schematic diagram of the device is displayed in Fig. 2a. Each nanowire was strained by an axial pressure. Because polarized nonflowing ions were distributed at both ends, a piezoelectric potential was generated inside the nanowire. When pressure was applied to taxels 45 and 46, an obvious and steady response could be observed, as shown in Fig. 2b. The introduction of the piezotronic effect into tactile sensors not only improved the resolution of devices but also extended some potential applications in intelligent systems. As shown in Fig. 2c, this group successfully realized an integrated 92 × 92 taxel strain-gated vertical piezotronic transistor (SGVPT) array. The two-dimensional current contour plot shows that the currents of the SGVPTs were very consistent. To prove the tactile sensitivity, this group pressed a die onto the device with a pressure of approximately 25 kPa. Different pressures showed different current values, and the two-terminal transistor array exhibited high resolution [44].

In addition, for visualization, the piezo-phototronic effect has been introduced to convert a force signal into an optical signal and reduce the resolution to the micron scale. Our group [45] developed a sensor array based on nanowire light-emitting diodes (LEDs) with a two-dimensional distribution of stress mapping. n-ZnO nanowires were patterned on a p-GaN substrate to form a pixeled light-emitting array, as shown in Fig. 2d. Each pixel consisted of a single n-ZnO nanowire/p-GaN LED. As shown in Fig. 2e, f, this device had an extremely high spatial resolution of 2.7 µm, which corresponded to a pixel density of 6350 dpi. The recovery time of the pressure sensor was fast, approximately 90 ms. Due to the piezo-phototronic effect, the local strain on the device determined the light emission density of each pixel. Afterwards, a “piezo” model on a sapphire substrate was used as a seal on the LED array to detect the distribution of two-dimensional strain. The light-emitting image for a strain of ~0.15% applied to the device clearly
Fig. 2. (Color online) High resolution of devices. SCVPT: schematic diagram of a three-terminal device and a two-terminal device (a), the response of taxels 45 and 46 to pressure (b), and tactile imaging by the fully integrated 92 × 92 SCVPT array (c). Reprinted with permission from Ref. [44], Copyright © 2013 AAAS. n-ZnO NW/p-GaN LED array based on the piezo-phototronic effect: schematic of the LED array exhibiting a pressure distribution (d), the resolution of the LED array was 2.7 μm (e), the recovery time of the pressure sensor was approximately 90 ms (f), and the LED array exhibited a stress distribution in the pattern “PIEZO” (g). Reprinted with permission from Ref. [45], Copyright © 2013 Springer Nature. (h–m) Flexible high-resolution sensors based on traditional and new materials: flexible ZnO NW/p-polymer LED array based on the piezo-phototronic effect (h, i), flexible Cds nanorod/organic hybrid LED array based on the piezo-phototronic effect (the inset bar is 200 nm) (j–l), and flexible ZnO NW/OLED array based on the piezotronic effect (m). Reprinted with permission from Refs. [46–48], Copyright © 2015 Wiley-VCH, © 2016, 2019 RSC, © 2017 American Chemical Society.
showed the word “piezo” with little crosstalk between adjacent pixels, as shown in Fig. 2g. Then, to further mimic human skin and enable adaption to intelligent systems, new materials, for example, ZnO, GaN and CdS NWs, were used to provide a way to simultaneously achieve flexibility and high resolution of sensors [49–51]. Based on the piezo-phototronic effect, our group [46] prepared a flexible LED array with ZnO NWs and p-polymer to achieve pressure mapping, as shown in Fig. 2h, i. Bao and co-workers [47] fabricated a PN junction, which consisted of CdS nanorods and organic layers grown on indium tin oxide (ITO) electrodes by the hydrothermal method, and they prepared an LED array based on the piezo-phototronic effect for mapping the pressure. A schematic diagram of the device is shown in Fig. 2j, k. The distribution of pressure had a high spatial resolution of 1.5 μm. The recombination of electron holes at the device interface determined its luminescence intensity. Fig. 2l shows an scanning electron microscope (SEM) image of the CdS nanorods; their tops were dipped into a poly(methyl methacrylate) (PMMA) layer. Bao et al. [48] designed an array that combines ZnO nanowires with organic LEDs. As shown in Fig. 2m, the light emission performance was determined by the organic luminescent layer. In addition, the emission was enhanced by modifying the energy band of the piezoelectric transistor and reducing the Schottky barrier between the electrode and semiconductor. Furthermore, the ZnO NW arrays regulated the spatial resolution of the device.

2.2. Stretchability

To simulate human skin as much as possible to meet the needs of intelligent systems, the stretchability of tactile sensors is critical. Stretchable conductors and other related electronic components have been extensively studied. A stretchable structure, an intrinsically stretchable conductor, and compounded conductive materials and elastomers are all widely used to implement device stretchability [52,53]. For instance, Guo and co-workers [54] developed a tactile sensor based on an electrode designed with argentum nanofibers (Ag NFs) and silk fibroin. The silk fibroin film exhibited excellent stretchability (>60%). Sun and co-workers [55] developed stretchable e-skin on the basis of precracked Ag nanowires. The resistance linearly varied with the density of cracks over a 30% strain range. Bao and co-workers [15] developed a transistor array based on an intrinsically stretchable polymer, with the design principle of the device illustrated in Fig. 3a. The device could work normally at 100% stretch, either parallel or perpendicular to the direction of current transmission. Since all the major thin films were based on SEBS (i.e., the dielectric, substrate, and 70% CONPHINE semiconductor), interfacial shear and vertical stress were effectively suppressed. In Fig. 3b and c, when a strain of 100% was exerted parallel to the channel direction, the electron mobility merely increased to 0.99 cm²/(V·s). Because of its high stretchability, the array could be comfortably attached to objects with naturally irregular surfaces and deformations (such as the palms of people) as a secondary skin, as shown in Fig. 3d. Polymer materials with intrinsically stretchable properties accelerated the development of stretchable tactile sensors [15].

Geometric design of devices is another main approach to achieve stretchability. Cao et al. [56] oxidized a specific part of the highly stretchable and conformable matrix network (SCMN) were connected by meandering wires. These meandering interconnects were vital to the implementation of high stretchability and expansibility. A customized pressure test platform was utilized to record the stretching process of the device, as shown in Fig. 3f and g. When the meandering wires were stretched to 800% of the original length, the tension and resistance of the meandering wires did not significantly change. An optical image of the stretched SCMN (scale bar: 2 cm) is shown in Fig. 3h. The sensor could be easily expanded in a specific direction to increase the sensing area [14].

Due to the importance of elastomeric materials and suitable structures for the stretchability of devices, a highly stretchable sensor array that combined Ag NFs with polydimethylsiloxane (PDMS) was developed. As shown in Fig. 3i, by electrospinning, the metal collector was charged to obtain polyvinyl alcohol NFs (PVA NFs) with different orientations, and then, Ag was deposited by magnetron sputtering to metallize the PVA NFs. PDMS acted as a substrate and an encapsulation layer. In this experiment, unidirectional, bidirectional, tridirectional and random Ag NFs were prepared. The effects of the orientation of Ag NFs and the angle of stretch were mainly studied. During the stretching process, although PDMS was used as a substrate to absorb stress through deformation because of its low stiffness, the unidirectional Ag NWs were cracked because of their high effective stiffness, so the device had many cracks at 0°, as shown in Fig. 3j. In Fig. 3k, for the tridirectional Ag NFs, as θmax (the maximum value of the angle between the tensile direction and one of the spinning orientations) increased, the change in resistance decreased. For the multidirectional Ag NF electrode, θmax was always 90°; therefore, this electrode had a better stretchable property, and the resistance only increased 10% after application of 100% strain, as shown in Fig. 3l. Obviously, using multi-oriented Ag NFs as conductive paths could greatly improve the stretchability of the electrodes [13].

2.3. Self-healing

Human skin can repair itself when exposed to external damage [57]. An ideal tactile sensor should demonstrate a similar capability of repeatable self-healing. A self-healing ability can greatly increase the lifetime of devices when they are damaged. In practical applications, sensors are required to repair damage at room temperature and repair repeated damage at the same location [58]. In recent years, many studies have been conducted on related aspects. The recombination of dynamic molecular bonds can restore the mechanical properties of devices. Self-repairable dynamic molecular bonds mainly include covalent bonds and supramolecular noncovalent bonds [59].

As a typical example, Wang and co-workers [17] introduced dynamic covalent imine bonds as reversible healing sites into a PDMS network. A schematic diagram is shown in Fig. 4a. By the Schiff base reaction, bis(amine)-terminated PDMS and 1,3,5-triformylbenzene generated reversible imine bonds as polymer crosslinker and self-healing points, which were cleaved upon damage and recovered after spontaneous healing. A thin film of a sprayed Ag NW network and a PEDOT film were used as conductors, which were placed on a prestretched PDMS substrate that was then released to produce a periodic wavy structure. Although the Ag-PEDOT film could not heal spontaneously, the healing of the polymer substrate involved fusion of H-PDMS to repair the crack, which exerted a traction force on the Ag-PEDOT film to re-establish electrical contact. The prestretched film had superior flexibility; therefore, the cutting process did not cause damage to the film. When the H-PDMS was completely cut by the blade, its ultimate stress returned to 94% of the initial state after 12 h (21 °C), as shown in Fig. 4b. In addition, Fig. 4c shows that after healing, the initial cleft between the buckled Ag-PEDOT films was clearly restored [17].
In addition to covalent adaptive bonds, supramolecular noncovalent bonds are another type of bond that can achieve self-healing. These bonds are involved in hydrogen bonding, metal–ligand coordination, π-π stacking, host–guest complexation, etc. [60–63]. Bao and co-workers [16] developed a sensor that can repair itself at room temperature. The self-healing mechanism mainly occurred through the reaction of oligomeric chains and μNi particles. As shown in Fig. 4d, a randomly branched network

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Fig. 3. (Color online) Stretchability of devices. (a) Schematic diagram of an intrinsically stretchable device. (b) Strain distribution in the array when stretched to 100% strain. (c) Mobility and threshold voltage during a parallel stretching cycle. (d) The device was attached to the palm of a hand and was consistent with it. Reprinted with permission from Ref. [15], Copyright © 2018 Springer Nature. (e) Schematic diagram of a stretchable structure device. (f) Relationship between the relative resistance of the device and the change in the force; the green portion is the area where the performance is stable, and the illustration is a schematic of the tensile test platform. (g) Stretchability of meandering wires; L/L₀ from 1 to 8. (h) Extended SCMN (scale: 2 cm); the illustration is an enlarged view of the device after stretching (scale: 5 mm). Reprinted with permission from Ref. [14], Copyright © 2018 Springer Nature. (i) Preparation process of a device in which a stretchable material is combined with a stretchable structure. (j) Deformation of unidirectional Ag NFs at different stretching angles. (k, l) ΔR/ΔR₀ of tridirectional Ag NFs (k) and random Ag NFs (l) varying with tensile strain. Reprinted with permission from Ref. [13], Copyright © 2018 Wiley-VCH.
was formed by linear and branched polymers, and hydrogen bonds were formed by urea groups at both ends of the branched polymer. The self-healing property of the supramolecular polymers was based on many weak hydrogen bonds, which were preferentially broken under mechanical damage. Through the dynamic association and dissociation of these “broken” hydrogen bonds at room temperature, the fracture surface underwent a passive healing process. In addition, the low glass transition temperature caused the polymer chains to rearrange, wet and diffuse at the fracture interface, which allowed completion of the self-healing process at ambient temperatures. Fig. 4e and f show the electrical healing process over time. When a light pressure was applied onto the wound for 15 s, the conductivity of the polymer was close to the initial value. After cutting and the self-healing process at the same position three times, the conductivity still recovered well. Fig. 4g shows the connection between the electrical conductor and LED. The accomplishment of the self-healing process of the electrical conductor was shown by the lighting of the LED [16].

Additionally, dual-strength dynamic metal-ligand coordination bonds, which can create attractive interactions, were embedded into PDMS to obtain the self-healing property. Fig. 4h shows the self-healing process of supramolecular bonds. The attractive metal-ligand coordination interactions as a broad molecular parameter can be used to impart desirable material properties by changing the metal ions, counterions, and ligands to adjust the bond strength. The bipyridine moiety has good coordination geometry with many transition metal ions, which can be used to direct self-assembly of polypeptides and construct metal–organic frameworks (MOFs) or molecular knots. Metal salts can be additional knobs for adjusting the crosslink density and polymer kinetics [64]. To test the self-healing performance, the relationship of strain and stress between the original membrane and the self-healing Zn

Fig. 4. (Color online) Self-healing of devices. (a) Self-healing was achieved by dynamic covalent imine bonds. Bis(amine)-terminated PDMS and 1,3,5-trimethylbenzene formed a reversible imine bond by the Schiff base reaction, which cleaved after injury and recovered during autonomous healing. (b) After being cut by a razor blade, the H-PDMS healed at different times under ambient conditions (21 °C). (c) Microscopic image of a new cut and the healing H-PDMS/Ag-PEDOT membrane at 12 h. Scale bar: 100 μm. Reprinted with permission from Ref. [17], Copyright © 2018 American Chemical Society. (d) Self-healing mechanism of supramolecular bonds (hydrogen bonds). The linear and branched polymers formed a random branching network, and the urea groups at the ends of the branched polymer formed the primary hydrogen bonds between the polymer chains. (e) During the healing process of the device, the resistance basically returned to the original value after 60 s. (f) Optical microscopy images of damaged samples and complete scar healing samples. Scale bar: 1 mm. (g) After self-healing of the conductive composite material, the LED was illuminated again. Reprinted with permission from Ref. [16], Copyright © 2012 Springer Nature. (h) Self-healing mechanism of supramolecular bonds (metal–ligand coordination bonds). Dynamic interaction between the metal cation Zn$^{2+}$, ligand and counter anion in the polymer system under mechanical stress. (i) Self-healing of the Zn(OTf)$_2$-PDMS polymer under ambient conditions. (j) Schematic diagram of the device after being cut and broken into two parts and subsequent self-healing. The broken part completely healed itself. Reprinted with permission from Ref. [59], Copyright © 2016 American Chemical Society.
et al. [77] developed a TENG based on the vertical contact-separation mode on an ITO-coated polyethylene naphthalate (PEN) substrate. The output voltage of the TENG is of the two friction layers, the authors obtained a high output voltage of 95 V and a short-circuit current of 180 μA.

The vertical contact-separation mode requires two electrodes connected by a load. However, in our daily life, some parts of a TENG involve movement, for example, the energy generated by the friction between a human walking and the earth; thus, connection by wires to electrodes is not feasible. To more conveniently collect mechanical energy in this case, a TENG with the single-electrode mode was proposed. Contact and separation occur between the upper charged object and the lower object, thus changing the distribution of the local electric field. To balance the potential, the electrode and ground exchange electrons with each other [79]. Zhao et al. [80] used carbonized polyaniline-nanofiber films (PAN NFs) as electrodes, embedded barium titanate nanoparticles (BTO NPs) into the NFs to produce the piezoelectric effect and then used PDMS as the encapsulation layer and triboelectric layer, as shown in Fig. 5a. SEM image of carbonized PAN/BTO NFs is shown in Fig. 5b. Based on the single-electrode mode, when a hand touched the PDMS, its surface was electrified due to friction. Meanwhile, the contact stress caused the BTO to deform, which resulted in the generation of a local piezoelectric potential that enhanced the potential of the surface. When the hand was removed, an equal amount of positive charge was generated on the PAN-C/BTO NF layer because of electrostatic induction, accompanied by the flow of charge, as illustrated in Fig. 5c. The open-circuit voltage and short-circuit current of the device were tested under different external pressures, as displayed in Fig. 5d. The electrical output ($R_\text{f} = 0.996$ for $V_{oc}$ or $R_\text{f} = 0.994$ for $I_{sc}$) and pressure had a good linear relationship from 0.15 to 25 N, which indicates that this type of device is reliable in pressure detection. A TENG based on the single-electrode mode was proposed by Fang et al. [81], with the fundamental structure exhibited in Fig. 5e. The device had a structure of PDMS-supported Ag nanowires, which could be used in intelligent systems and artificial skin. The contact-separation between PDMS and a finger was used to convert the friction signal into an electrical signal. The single-electrode TENG had a high electrical output. Fig. 5f, g shows that the peak value of the open-circuit voltage was approximately 70 V, and the short-circuit current density reached 1.5 mA/m² [81].

2.5. Biodegradability

With the increasing demand for intelligent devices, devices with novel performance must be developed to solve the problem of electronic waste. Therefore, the study of transient electronics has great significance. Furthermore, in the medical field, sensors have been widely used as medical devices to monitor health or treat some diseases, such as arrhythmia and bradycardia [82]. For the treatment of diseases, usually, the devices must work for a period of time. Therefore, when devices are implanted in vivo, biodegradability is extremely important. Good biodegradability can avoid secondary surgery for patients, which makes the treatment of diseases safer, simpler and more reliable [83].

To reach these goals, inorganic materials were first applied. In particular, silicon-based electronic devices play representative roles because of their ability to hydrolyze under physiological conditions. Biodegradable tactile sensors can be composed of silicon nanofilms and inorganic films, such as magnesium or magnesium oxide [20]. For example, Hwang and co-workers [84] developed a silicon-based strategy to fully form transient circuit components and circuits, relying on a combination of a particular type of silicon
on insulator (SOI) wafer and transfer printing technology. The materials of the device were all water-soluble, including silicon, siloxane, magnesium, magnesium and silk. And their applications in the manufacture of various electronic components and simple logic gates were demonstrated and their transient characteristics were studied. Canan et al. [85] put forward the concrete design and manufacturing plan of ZnO thin film optical transistor and mechanical energy harvesters (also can be used as a strain gauge) and introduced several electronic devices based on ZnO. The key features of these could be completely dissolved in the water or biofluids. Meanwhile, the kinetics of dissolution and the ability to control dissolution using materials and design choices are studied in detail. However, tactile sensors based on inorganic materials are rigid and require high technology and high cost. Later, to increase the flexibility of tactile sensors, miniaturize them and reduce the manufacturing cost, breakthroughs were made in the field of organic materials. The preparation process of organic electronics is more environmentally friendly. Some biodegradable small molecules have been reported, such as indigo derivatives and β-carotene [21,86,87]. Wang and Li groups [88] developed a biodegradable TENG (BN-TENG) based on natural materials to treat heart disease. The structure of the device is shown in Fig. 6a. In this experiment, all biodegradable materials came from nature, including cellulose, rice paper, silk fibroin, egg white and chitin. The BN-TENG included polymers (NBPs) and Mg electrodes. Any two different NBPs could be made from the five natural materials mentioned above for use as the triboelectric layer, and an ultrathin Mg film was used as the back electrode. The surface of the triboelectric layer was treated by inductively coupled plasma (ICP) to form an array of nanostructures to improve the effective contact area and contact electrification. The biodegradability performance was characterized \textit{in vitro} and \textit{in vivo}. Fig. 6b (top) shows the biodegradation process \textit{in vitro}. BN-TENGs encapsulated with silk fibroin were placed in phosphate buffer solution (PBS). The structure was intact on the first day. Over time, the skeleton of the polymer was broken, autocatalytic hydrolysis occurred, and finally, the
Fig. 6. (Color online) (a) Schematic diagram of a BN-TENG based on biodegradable organic polymers derived from nature. Lower and upper scale bars: 5 and 1 μm. (b) Degree of degradation of the device after different times in vitro (top) and in vivo (bottom). Reprinted with permission from Ref. [88], Copyright © 2018 Wiley-VCH. (c) Schematic diagram of a device prepared from a synthetic biodegradable organic polymer. Scale bar: 500 μm. (d) Degradation process of the device in vivo. (e) Degradation process of the device in vitro. Reprinted with permission from Ref. [89], Copyright © 2018 Elsevier. (f) Schematic diagram of a biodegradable material and structural diagram of the device using a reversible imine bond. (g) Process of device degradation. (h) Ideal cycle diagram for the synthesis, fabrication, and decomposition of transient polymer electronics. Reprinted with permission from Ref. [23], Copyright © 2017 National Academy of Sciences.
whole structure was dissolved in PBS. As shown in Fig. 6b (bottom), BN-TENGs encapsulated with silk fibroin were placed in SD rats. The biodegradation process was roughly the same as that in vitro, and the cracked parts were degraded and absorbed by the SD rat. However, the biodegradation process could not be controlled from the outside. To achieve regulation of the biodegradation process, a tunable biodegradable implantable TENG (BD-iTENG) was fabricated by Wang and co-workers [89], and the biodegradation process was studied in vivo and in vitro. Fig. 6c shows that the ability of the BD-iTENG to respond to near-infrared (NIR) irradiation derived from the poly(lactic acid-glycolic acid copolymer (PLGA) membrane doped with gold (Au) nanorods in the BD-iTENG, so we could use some external conditions to control the biodegradation process. The BD-iTENG included three parts: (1) an array of hemispherical structures was deposited with Au as both a triboelectric layer and a bottom electrode; (2) a biodegradable polymer layer was deposited as another triboelectric layer, on which Mg was deposited as an upper electrode; and (3) another biodegradable polymer layer doped with Au nanorods acted as a substrate. Fig. 6e shows that the BD-iTENG was placed in PBS at 37 °C for 35 d. In the absence of NIR treatment, a majority of the PLGA-iTENG was hydrolytically degraded by PBS after 35 days. When the BD-iTENG device was treated by NIR, the degradation rate was significantly increased. After that, PLGA-iTENGs treated by NIR were implanted into the body of SD rats. Fig. 6d shows that the PLGA-iTENGs gradually degraded into fragments within 28 d. After one NIR treatment, the voltage decreased to 0 at the 24th hour.

In addition to some natural materials and the synthetic materials mentioned above that have biodegradability, conjugated polymers have been developed to achieve biodegradability of tactile sensors for intelligent systems. Conjugated polymers are considered candidates for field-effect transistors (FETs) due to their good flexibility, simple preparation techniques, and high carrier mobilities [90]. Bao's group [23] demonstrated a thin-film transistor with a semiconductor polymer, which was composed of reversible imine bonds and provided building blocks. In addition, this transistor easily decomposed under mild acidic conditions. Fig. 6h shows several key processes of the “ideal cycle”. The synthesis of semiconducting polymers from substances in nature and their eventual decomposition in the environment are anticipated. In the study by Bao et al., the imine bond was used as a “reversible conjugated linker”, as illustrated in Fig. 6f. To realize biodegradability of the whole electronic device, iron (Fe) was introduced as a gate electrode, and a source-drain electrode of the polymer and PEDDP-PD was synthesized via the condensation reaction. The degradation process of the flexible device is shown in Fig. 6g. The Fe electrode quickly degraded within 1 h under certain conditions, and the conjugated polymer, cellulose substrate and alumina could be fully degraded within 30 d. In practical applications, the use of suitable decomposable encapsulation materials could potentially enable control of the rate of device degradation.

3. Applications in intelligent systems

Due to the high performance of tactile sensors, they are being applied to intelligent systems increasingly frequently. In this section, we introduce three application areas and the latest researches in intelligent systems. Wearable devices offer broad prospects for the development of mobile medical and visualization devices [91]. In the field of biomedicine, special function devices provide ideas for the design of disease monitoring systems in vivo and in vitro, as well as antibacterial platforms. The development of electronic signature systems makes data collection and management more convenient and guarantees the security of personal information [92]. The study of electronic skin not only enables the realization of more intelligent robots but also has important significance in medical treatment.

3.1. Wearable devices

The materials used to fabricate wearable devices must meet the requirements of being flexible, lightweight, breathable and bio-compatible and having some special function. Wearable devices not only provide an approach to real-time health monitoring and personal communication but also have convenient portability [93]. Consequently, significant efforts are being made to develop wearable devices.

Primarily, wearable devices are applied in real-time health monitoring. Li and co-workers [94] developed a flexible nanogenerator (NG) that acted as a wearable self-powered breathing sensor for real-time monitoring of human breathing. The poly(vinylidene fluoride) (PVDF) film prepared by electrospinning acted as a piezoelectric layer due to its high piezoelectric voltage constant. Au was deposited as the top and bottom electrodes on the two sides of the PVDF film. The piezoelectric NG was placed on the chest of a man. Using the platform of the piezoelectric active sensor based on the PVDF piezoelectric film (PEAS) and a Biopac MP150 physiological signal recording system, different frequency respiratory signals were measured. The PEAS and Biopac MP150 were highly consistent at different respiratory rates, and these breathing curves contained detailed information about the dynamics of the respiratory process. The PEAS was used to detect vibration of the vocal cords. No signal was observed when no sound was heard. When ‘P, I, E, Z, O’ were spoken, regular high frequency signals appeared. Different letters had different piezoelectric signals. Therefore, the PEAS for use in quantitative diagnostic techniques might evolve as a wearable and zero-power-consumption device in the future.

Smart textiles can change depending on the environment, such as temperature and color. A communicative textile with a large area can be made and easily integrated into a human. Among smart textiles, light-emitting textiles have aroused great interest because of their high visibility, security and fashionability [95]. One of the most typical light-emitting textiles is realized by weaving optical fibers according to a specific pattern and incorporating them into the textile. Jeong and co-workers [96] designed a mechanoluminescent (ML) textile that used the movement of a human as the power source to drive the optical fibers to emit light. Stretching of a muscle could cause mechanical deformation of the fibers. As shown in Fig. 7a, ZnS particles were doped with metal ions and embedded in PDMS + ZnS, and a cross-shaped fiber frame was used to make the contact area with PDMS + ZnS larger, which improved the adhesion between the materials. At the same time, the use of an organofunctional silane coupling-based primer improved the reactivity and adhesion of the fibers. After that, the fiber coated by PDMS + ZnS was covered with Si(adh) to fill the gaps or defects in PDMS + ZnS. The PDMS + ZnS was surrounded and 3D localized by three contact points. The authors used a shuttle to tightly bind the fabrics together. The Si(adh) was transparent, so the edges of the fibers indicated that Si(adh) adhered well to the fibers and PDMS + ZnS. Fig. 7b shows a photograph of the weaving process of ML fibers. To verify the ML properties, we applied tension to the ML fabric and observed green light, as shown in Fig. 7c.

In addition to ML fibers, electroluminescent fibers can also be integrated into fabrics for the preparation of multifunctional wearable electronic devices [98]. A novel electroluminescent textile display obtained by a continuous one-step extrusion process was demonstrated by Zhang and co-workers [97], as shown in Fig. 7d. A combination of ZnS powder and a silicone elastomer formed the electroluminescent material. The silicone elastomer acted as both a protecting layer and a dielectric layer. The design of the
two elliptical hydrogel electrodes increased their face-to-face area so that they could emit stronger light, as shown in Fig. 7e. PVA and poly(ethylene oxide) (PEO) were used as internal conducting electrodes. Due to the unique fiber structure, the super-stretchable electroluminescent fibers (SEFs) could emit light in all directions, but the maximum luminance was at angles of 0° and 180°, as shown in Fig. 7f. When the length of the SEFs reached the meter level, the change of luminance along the axial direction was less than 5%. Although green light was dominant, other colors, such as yellow and blue, could also be achieved by selecting the desired ZnS powder. The SEFs could be stretched to 800%. Moreover, the fibers also had extremely high stretchability and could be stretched up to 800%. As shown in Fig. 7g, the textiles could be connected to a computer. When different numbers were entered into the computer, different numbers were displayed on the textile. Afterwards, using the integrated platform, a brain-interfaced camouflage system was constructed, in which the color of the electroluminescent textile could be determined by the decoded neuron, and when the ambient light changed from green to blue, the textile could quickly respond with discoloration, as shown in Fig. 7h.

3.2. Biomedical field

Biomedical tactile sensors are mainly composed of two key parts: the part for receiving and generating the signal of tactile sensors and the hardware instruments of the devices for converting the physical signal. The former mainly include electrical and optical detection components, such as current/potential measuring electrodes, piezoelectric crystals and FETs [99–101]. The emergence of various new materials and new technologies has enabled biomedical tactile sensors to be miniaturized, multifunctionalized, integrated and made intelligent. At present, thermosensitive biomedical sensors, FET biomedical sensors, piezoelectric biomedical sensors, triboelectric biomedical sensors and photosensitive biosensors have been widely used in biomedical research [102].

Among them, triboelectric biomedical tactile sensors have gained great attention because of their self-powered capability, low power consumption and high sensitivity. A self-powered ultra-sensitive pulse sensor (SUPS) for intelligent mobile diagnosis of cardiovascular disease was developed by Wang and co-workers [103]. The design principle of the device was the coupling effect of contact electrification and electrostatic induction. A schematic diagram of the device is shown in Fig. 8a. A Kapton thin film and an ultrathin copper (Cu) layer with nanostructures were used as two triboelectric layers. The ultrathin Cu layer and another Cu layer plated on the back of the Kapton film were used as electrodes, and PDMS was used as an encapsulation layer. The device adopted concave structures to ensure an excellent correspondence between the force and the electrical signal. Heart rate variability (HRV) is a convincing predictor of cardiovascular disease, and the geometry of the HRV Poincare plot was used to clinically distinguish the health of subjects. Fig. 8b, c show the Poincare plots constructed...
from the radial artery pulse of a healthy person and a patient with coronary heart disease (CHD). The Poincare plot of a healthy person was comet shaped or elliptical; however, a Poincare plot with shorter width and length indicated that the patient had CHD, proving that the SUPS was effective in the diagnosis of cardiovascular disease. The system is expected to find application in intelligent mobile diagnostic systems.

To prepare biomedical devices that could be implanted into the body, Wang and Li made some efforts in this field. For patients with impaired cardiac function, the changes in endocardial pressure have a certain determinant role [106]. Based on a TENG, a flexible, miniaturized, self-powered endocardial pressure sensor (SEPS) was fabricated, which was minimally invasively implanted with surgical catheters. As shown in Fig. 8d, polytetrafluoroethylene (PTFE) etched by ICP was used as a triboelectric layer, an aluminum (Al) foil was used as another triboelectric layer, and a 3D ethylene–vinyl acetate (EVA) copolymer film acted as a barrier to ensure contact-separation between both triboelectric layers. The nano-PTFE surface was modified by corona discharge to achieve a high surface charge density and improve the sensitivity and output performance of the SEPS. To simulate the role of the SEPS in biomedical real-time monitoring, the authors chose male...
adult Yorkshire pigs (40 kg) as the animal model, as shown in Fig. 8e. They combined the miniature SEPS with a polyvinylchloride (PVC) retractable catheter to construct a delivery system to achieve minimally invasive implantation. The working signals of electrocardiography (ECG), the femoral arterial pressure (FAP), and the SEPS were compared in the resting, waking, and active states, as shown in Fig. 8f. In the resting state, the device had little effect on the stability of the cardiac functionality. In the active state, the SEPS responded to endocardial pressure, and the peaks of the SEPS outputs showed slight fluctuations with the systolic FAP (arrows), indicating that the endocardial pressure remained high. Furthermore, the device still had a high sensitivity to small changes [104].

In many clinical applications, antibacterial properties are also of great significance. Physiologically, electrical interactions cause changes in bacterial morphology. Therefore, Chu and Li group [105] were inspired to design light-independent antibacterial materials and introduced other sterilization measures, such as using electrically modified biological materials to supplement light irradiation. Titanium metal foil was placed in NH4F-ethylene glycol solution for electrochemical anodization to prepare titania nanotubes, and then, the obtained carbon nanotubes were annealed in argon (Ar) to prepare ordered carbon-doped capacitive titanium dioxide nanotubes (TNT-C), as shown in Fig. 8g. Scanning transmission electron microscopy (STEM) and electron energy loss spectroscopy (EELS) demonstrated that Carbonium (C) was evenly distributed across the entire tube wall, as shown in Fig. 8h. An impressed current was applied to the TNT-C to evaluate the bactericidal ability. In different TNT-C samples, TNT-C-15 with the highest capacitance had the highest inhibition rate, which indicated that the antibacterial property of the TNT-C sample was related to the capacitance and the thickness of the bacterial membrane after charging, as shown in Fig. 8i.

### 3.3. Man–machine interaction and intelligent robots

Practical breakthroughs in advanced tactile sensor technologies, such as image recognition, voice recognition, pressure recognition and temperature recognition technologies, have led to significant improvements in man–machine interaction and the perceptual capabilities of robots [107]. Electronic devices and robots are beginning to enter a new era of intelligence.

Among them, the man–machine interaction system realizes the transformation of information into a human acceptable form. A wafer-scale flexible pressure sensor was invented that achieved secure signature collection by recording the signer’s handwritten pattern and signing habits. The device used ML materials (ZnS: Mn particles), which were bonded with polyethylene terephthalate (PET) to form a sandwich structure, as shown in Fig. 9a. In the single-point dynamic pressure recording and two-dimensional planar pressure mapping mode, the devices responded immediately to the stimulation through the mechanoluminescence process, which converted the pressure signal into an optical signal. Fig. 9b shows the image acquisition process and the information processing system. As shown in Fig. 9c, by pressing a stamp onto the pressure sensor matrix (PSM) device, a “rose-shaped” pressure profile on a two-dimensional plane was obtained. Fig. 9d shows the recorded results of the signing habits of several signers. Through the signer’s handwritten “piezo” and the corresponding ML intensity distribution, differences in morphology could be clearly observed. According to the image, we could also see personal handwriting habits, such as the pressure on each pixel and the handwriting speed [108]. In addition, the choice of different materials allowed the device to emit light of different colors. Piezoelectric material CaZnOS:Er3+ particles exhibit mechanoluminescence and upconversion luminescence properties. The device was a flexible film made of two polyethylene terephthalate substrates with CaZnO:Er3+ particles coated between them, and green light could be observed [110]. Lanthanide-ion-doped CaZnOS:Sm3+ emitted red light upon stimulation by a dynamic mechanical stress [111].

Through the coupling effect of photoluminescence and triboelectrification, a stretchable intelligent skin for tactile perception and gesture sensing as developed. Fig. 9e shows a schematic diagram of the tactile sensor. A flexible tightly bonded microcracked metal film with an aggregation-induced emission (AIE) compound served as an active substrate. The microcracks acted as “gates” to regulate the exposed area of the AIE-active substrate and the PL intensity from the stretchable triboelectric-photonic smart skin (STPS), as shown in Fig. 9f. The Ag NW network in the middle served as a stretchable conductive layer. The AIE compound, bis (4-phenothiazinophenyl) sulfone, acted as a solid intense fluorescent emitter. A crack occurred at the position where the finger was bent, thereby causing light to be emitted. Furthermore, the authors integrated the STPS onto the five fingers of a robot. As shown in Fig. 9g, when the gesture “OK” was made, the 3D distribution of the light signal and the luminescent photograph indicated the degree of bending of each finger [109].

Furthermore, the development of intelligent systems enables robots to have auditory sense and tactile sense. A circular-type single-channel self-powered triboelectric auditory sensor (TAS) was designed, which was used to construct an electronic auditory system and an external hearing aid for robots. A schematic diagram of the TAS is shown in Fig. 10a. The TAS was a circular device consisting of fluorinated ethylene propylene (FEP) plated with a top electrode and comprising a plurality of channels, as well as a Kapton film with a bottom electrode. A gap was formed in the middle so that the inside could freely vibrate. With the application of the TAS, sound could be used to turn on a console light and to trigger an antitheft system, as shown in Fig. 10b. Due to the structure of the triboelectric cochlea (TEC) device, it could be easily integrated into the artificial ears of robots. Fig. 10c shows the sound wave signal of original music and recorded music. The graph shows that the two sound wave signals were almost identical. Moreover, the voice recognition capabilities of the robot are demonstrated in Fig. 10d, e, which proves the feasibility of using the TAS in a robotic auditory system [112].

To achieve perception of multiple stimuli by a robot, Wang and Pan groups [14] presented a stretchable tactile sensor network of island-bridge structures, which was used to simulate human somatosensory systems, as shown in Fig. 10f. This network could detect temperature, relative humidity, magnetic field, in-plane strain, ultraviolet light, proximity and pressure. The perception of temperature and pressure is an important feature of human skin, so smart prosthetic hands have been designed that can be used to feel pressure and temperature. Fig. 10g shows the distribution of five tactile pressure sensors and tactile a temperature sensor on an intelligent prosthetic hand. Fig. 10h shows the different responses of the smart prosthetic hand when grasping and releasing objects. At the same time, when the smart prosthetic hand grabbed a cup, it would make different inferences for different temperatures.

### 4. Conclusion and perspective

In this review, we introduced the rapid development of material technology, flexible electronics, and nanotechnology in the field of tactile sensors and their application in intelligent systems. Recent studies have demonstrated improvements in the resolution, sensitivity, and other properties of devices. Meanwhile, the designs of devices have become more reasonable and mature to provide multifunctionality, such as stretchability, self-healing, self-powered...
capability and biodegradability. Thus, the development of tactile sensors provides a platform for wearable devices, biomedical systems, man–machine interaction systems and intelligent robots, which pushes the development of intelligent systems.

However, tactile sensors still face enormous challenges in practical applications. First, the performance of tactile sensors, such as the sensitivity and flexibility, is still the main research field. With the application of surface micromachining technology, researchers can develop tactile sensors from tactile arrays to high-density arrays to increase the resolution and sensitivity. In addition to the development tendency mentioned above, the use of piezoelectric nanowires can realize high resolution and low crosstalk between pixels, and on this basis, the introduction of TENGs can enable a device with low power consumption and high sensitivity.

Elastomeric materials and stretchable structures have also successfully expanded the effective sensing area of devices. PENGs and TENGs have been proposed to enable devices with a self-powered capability. With the study of self-healing materials and biodegradable materials, tactile sensors are becoming increasingly widely used in intelligent systems and provide a simpler and safer development direction for wearable devices, implantable devices and so on. Meeting these challenges will bring new opportunities. Second, sensors undergo repeated deformation during actual use, and the deformations will affect the performance of devices. Therefore, the detection accuracy and lifetimes of devices are seriously reduced. Researchers can make greater efforts in realizing advanced materials and suitable structures to improve the durability of devices. At the same time, the development of packaging
materials and packaging technology is another important way to improve the stability and durability of devices. Third, in practical applications, multiple stimuli tend to occur in the same place at the same time. To achieve the detection of all stimuli, decoupling of crosstalk signals of tactile sensors is required. The simultaneous collection of multiple signals with no crosstalk is important and is of great significance to developing new tactile sensors that can be applied in intelligent systems. In addition, the integration of the sensing system also imposes requirements on the components. Each component of tactile sensors in an intelligent system must have mechanical, optical, electrical and thermal property matching. Meeting these challenges will bring new opportunities. The tactile sensors used in intelligent systems will be flexible, miniature, and multifunctional.

Conflict of interest

The authors declare that they have no conflict of interest.
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Author contributions

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References


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