

Research Progress of Flexible Wearable Electronic Device

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Abstract: With the development of the times, people's lives are becoming more intelligent. Therefore, many intelligent terminals are widely used. Wearable electronic devices are at the forefront of research in the world today (due to their interactive design, Workers can easily know what they are in through the devices they wear. This allows them to respond in a timely manner. Therefore, the research of wearable electronic devices has become a new research direction. In this paper, the research progress of flexible wearable electronic devices in recent years is reviewed.

Keywords: wearables, flexible, medical, manufacturing, overview

1. Introduction

Electronic sensor is one of the core components of electronic wearable devices, which can affect the function and development of wearable devices. Therefore, the preparation of electronic sensors is particularly important. Making wearable flexible electronic sensors is more demanding than making ordinary electronic sensors [1]. For example, flexible electronic sensors need to be lighter, more portable and softer than ordinary electronic sensors. Better performance and high integration. In recent years, significant progress has been made in the research of flexible wearable sensors. Nowadays, flexible wearable electronic sensors can use electronic skin to transmit skin tactile information to the brain. Three-dimensional microelectrodes are used to control the artificial limb by the cerebral cortex, and cochlear implants are used to restore the hearing of patients.[2]

Wearable sensors need to have a positive impact on the human body. Such as a body temperature detector, an artificial electronic skin, a heart detector and the like. At present, researchers have developed a variety of ways to use sensors in the wearable field. Most of these studies are from a medical point of view. The temperature sensor can be used in the wearable field.[3] Generally, the body surface temperature of normal human body is maintained at about 32 degrees Celsius, while the core temperature of human body is about 37 degrees Celsius. Therefore, whether the temperature of the human body is normal or not can be effectively detected through the temperature sensor so as to judge the physiological state of the human body [4], O as to give corresponding suggestions to the wearer. In addition, pressure sensors also play a great role in the wearable field. It can be used as a device for sensing external pressure or converting human body signals into electrical signals in flexible skin, Therefore, it has been continuously concerned and studied in the fields of artificial electronic skin, wearable and health detection. In the field of health detection, pressure sensors are mainly used to detect pressure from the inside of the human body. For example, the blood pressure and pulse of the human body are detected.[5]

2. Signal conversion mechanism of electronic sensor

The typical signal conversion mechanisms of wearable flexible electronic sensors are piezoresistive, piezoelectric and capacitive. There are other conversion mechanisms that have not yet been fully developed, such as triboluminescence and force generation.

Piezoresistive sensor can convert the external force into the change of resistance, and then it is convenient to detect the change of external force indirectly with an electrical test system [6]. The change of conductive path between conductive materials is a common mechanism to obtain piezoresistive sensing signals.[7] This type of sensor is widely used due to its simple device and signal readout mechanism. Variation of the conductive path between conductive substances is a common mechanism for obtaining piezoresistive sensing signals, the change in contact resistance between the conductive substance and the conductive elastic composite material is proportional to the square root of the applied pressure, Piezoresistive sensors are usually used to detect small pressures and expand the detection range [8].

Cheng et al. Developed a simple and practical piezoresistive sensor with high sensitivity. Thin layers of gold nanowires and electrode arrays are fabricated on flexible substrates. The device has a wide detection range from 13 to 50000 Pa. In order to enhance the sensitivity, a scanning of the contact force is realized [9], Bao et al. made an electronic skin that can transmit tactile information to the brain by using a piezoresistive sensor with a conical microstructure. However, if the piezoresistive sensor is to achieve high sensitivity, it needs to be at the expense of high resistance.[10]

Recently, Zhang and others have developed a nano-graphene piezoresistive film [11], which improves sensitivity and reduces power consumption. In order to achieve a differentiated resistance response, Professor Qian Xin et al [12]. Assembled a micron-scale curve array piezoresistive sensor with adjustable amplitude and period by using silicon pillar induced printing method. It maintains a stable real-time response at 1000 cycles of 5% tension-relaxation. Silver nanoparticles and conductive polymers were successfully assembled in the experiment. This method is universal because it is based on the principles of liquid bridge contraction between silicon pillars and nanoparticle self-assembly.

2.1. Capacitance sensor

Conventional capacitive sensors detect different forces, such as pressure, shear, etc., by varying the facing area S and the parallel plate spacing d . The main advantage of capacitive sensor is its strong sensitivity to force, which can achieve low energy consumption to detect small static force. Bao et al. [9] Prepared a capacitive transparent and stretchable carbon nanotube sensor on an elastic substrate, which responds to both pressure and tension.

2.2. Piezoelectric materials

Piezoelectric materials are special materials that can generate electric charges under mechanical pressure. This piezoelectricity is caused by the presence of an electric dipole moment. The electric dipole moment is obtained by a non-centrosymmetric crystal structure deformation of the orientation, or a porous electret in which charges persist in the pores. [13]]Piezoelectric materials with high sensitivity, fast response and high piezoelectric coefficient are widely used in sensors that convert pressure into electrical signals. Piezoelectric inorganics are typical materials with high piezoelectric coefficient and low flexibility. Piezoelectric polymers are just the opposite. In order to explore the flexible pressure sensor with high piezoelectric coefficient, a series of methods have been tried. Includes constructing a piezoelectric inorganic thin film on a flexible substrate,[14] Piezoelectric polymers or inorganic/polymer composites are used and stable piezoelectric electrets are constructed. Lately, Nanowires and nanobelts with good piezoelectric properties and mechanical stability have attracted international attention to integrated high-resolution sensing array sensors [15].

3. The manufacture of flexible wearable electronic devices

3.1. Material selection of flexible electronic sensor

The first is the choice of a flexible substrate. Polydimethylsiloxane (PDMS) is widely used, its advantages include convenience [16], chemical stability, transparency and thermal stability. Especially under ultraviolet light, the distinct characteristics of adhesion area and non-adhesion area make it easy to adhere electronic materials on its surface. In addition, in the selection of flexible substrates, PET has good creep and fatigue resistance, and is less affected by temperature changes. It can be used for a long time at 55 ~ 60 °C. For low temperature applications, PI is a polymer with imide rings in the main chain, and its glass transition temperature is about 404 °C. It can withstand the extremely low temperature of -269 °C.[17]

In order to meet the demand of environmental friendliness, flexible biodegradable substrates are also one of the research hotspots. Aliphatic-aromatic random copolyester (ECOFLEX) is a biodegradable plastic with high elongation at break, has good ductility and temperature resistance.[18] Natural rubber (NR) [19] is one of the most important polymers produced by plants, which has high elasticity and elongation at break. It is a hot base material for preparing degradable sensors. In addition, thermoplastic polyurethane (TPU) has good processability and cold resistance, and can still maintain flexibility at -35 degree C° can be degraded under certain temperature and environment.[20]

For electrodes and wires, metal materials are generally used. For modern printing processes, conductive nano-inks, including nanoparticles and nanowires, are often used as conductive materials. In addition to good electrical conductivity, metal nanoparticles can also be sintered into thin films or wires. Park et al. developed a circuit for mass production of silver nanoparticle-coated rubber fibers by electrospinning.[21] Conductivity reaches $2200 \text{ S} \cdot \text{cm}^{-1}$ at 100% tension. In addition, liquid metal is a new generation of functional materials with both fluidity and metal conductivity at room temperature [22]. Compare with mercury, that low vapor pressure gallium-indium alloy has the advantages of stability, non-toxicity, low Mel point and the like, It shows great potential in flexible conductive materials. Liu Hong et al.[23] Dispersed magnetic nickel particles in gallium-indium alloy. And a moving magnetic field is applied to enable the magnetic particles to aggregate inside the liquid metal and generate the capillary phenomenon, Direct patterning can be achieved on irregular surfaces and superhydrophobic metal substrates. They believe that nickel particles are concentrated in the liquid metal under the action of magnetic field, and there are small gaps between the particles to produce capillary force. [24] The adhesion between the liquid metal and the substrate is enhanced. The method has advantages and limitations, and solves the problem that the existing patterning technology is difficult to realize the application of devices with complex morphologies, However, the sensitivity of the sensor at high strain is low.[25]

In addition to metal materials, conductive hydrogel electrodes are also a good choice. Gel [26] is a hydrophilic polymer material with three-dimensional network crosslinking structure, and its Young's modulus matches human skin tissue. Has good biocompatibility, strong adhesive force and tensile resistance. The conductive polymer hydrogel is a composite biomaterial formed by combining a polymer with conductive activity and a hydrogel, beside keeping that original characteristic of the hydrogel, the hydrogel also has large specific surface area, adjustable conductivity and high ion and electron mobility, is an ideal material for preparing a wearable electrode. Han et al. [27] doped polyaniline and carbon nanofibers (PANI-CNF) into polyvinyl alcohol (PVA) matrix. Since the good dispersion behavior of PANI-CNF in PVA network provides an excellent 3D interconnection path for electron transport, The conductivity of the prepared PANI-CNF-PV a hydrogel electrode is as high as $(5.19 \pm 0.07) \text{ S/m}$. Khademhosseini et al. [28] Used silver nanowires as conductive filler and methacryloylated sodium alginate as organic binder. A printable conductive hydrogel electrode was prepared, which benefited from the role of double crosslinking structure. The electrode has a

conductivity as high as 1.54×10^5 S/m. [29] Although the conductivity of the conductive composite can be improved by adding the conductive material into the hydrogel, due to the volume load of the high-stiffness material, [30] the mechanical properties of the hydrogel are inevitably impaired. Chen Xiaodong et al. infiltrated the hydrogel precursor into the porous TPU network. A hydrogel elastomer hybrid with a mechanical interlocking structure is prepared by a thermal curing method, then the electrode is combined with a nano-gold film by a thermal evaporation method, and the electrode can be attached to the skin for electrophysiological signal recording. [31] In addition, some wearable electrode materials improve conductivity by increasing the content of conductive fillers. [32] However, with the increase of conductive filler content, its conductivity and flexibility are difficult to compromise, which can not meet the application of digital circuits. Majidi et al. Added silver flakes as a filler to a polyacrylamide alginate hydrogel, The high valence cations are removed and subjected to a crosslinking reaction.[33] Then the composite hydrogel is kept stand to remove partial water, and the volume of the composite hydrogel is reduced along with the evaporation of the water to press a silver sheet, And then overlap with each other to form a seepage network, so that that conductivity of the electrode can reach 374S/cm. In addition, the flexibility of the hydrogel electrode is less than 10 kPa, which is comparable to that of human adipose tissue.[34] Most of the flexible electrodes based on hydrogel have poor toughness and durability, so efforts should be made to effectively prolong the service life of hydrogel electrodes. Such as improving the mechanical properties of the hydrogel, especially the toughness, or endowing the hydrogel with self-healing ability. [35] In addition, the variable temperature environment has a great influence on the conductive hydrogel, and its performance depends largely on the working temperature.[36] Clarifying the microscopic physical mechanism of the effect of temperature on device charge transport has become the research focus of the next generation of conductive hydrogel-based electrodes.

Some inorganic semiconductors also play a role in the preparation of flexible wearable electronic devices. Inorganic semiconductor materials such as ZnO and ZnS have excellent piezoelectric properties, It shows broad application prospects in the field of wearable flexible electronic sensors. [37]A flexible pressure sensor based on the direct conversion of mechanical energy into an optical signal was developed. This matrix makes use of the mechanoluminescence properties of ZnS:[38] Mn particles. The core of the mechanoluminescence is the photon emission induced by the piezoelectric effect. The electronic energy band of the piezoelectric ZnS generates a voltage-voltage effect under the action of pressure to generate inclination,[39] so that the excitation of Mn^{2+} can be promoted, The following de-excitation process emits yellow light (around 580 nm).A kind of sensor with fast response (response time less than 10 ms is obtained by this conversion process.[40] Through a top-down lithography process, the spatial resolution of the sensor can reach 100 μm . The sensor can record the dynamic pressure of single point slip. It can be used to identify the signer's handwriting and to scan two-dimensional plane pressure distribution by obtaining the emission intensity curve in real time. [41] All of these characteristics make inorganic. Conductor materials have become one of the most potential candidates in the field of fast response and high-resolution pressure sensor materials in the future.

Carbon materials may also be used in the preparation of flexible electronic devices. Carbon nanotube, graphene and MXene in carbon-based materials have excellent mechanical and electrical properties,It has great potential in the preparation of sensor functional layer materials, which can be divided into fiber assembly, film assembly and block assembly according to the assembly mode.[42] The carbon nanotube has the advantages of high crystallinity, good electrical conductivity, large specific surface area, controllable micropore size through a synthesis process, The specific surface utilization rate can reach 100%. Graphene has the characteristics of lightness, transparency, good electrical and thermal conductivity, etc.[43]

3.2. Preparation theory

For implementing a flexible sensor, the thickness of the substrate can be reduced so that the flexibility of the device can be increased. However, this method has a big problem, that is, it needs a nearly flat substrate surface. This results in a very limited shape of the substrate. Thus, a stretchable electronic device is proposed. Because this stretchable electronic device can be fully adhered to complex and uneven surfaces.

Currently, there are generally two strategies to achieve stretchability of wearable sensors. The first approach is to bond a thin conductive material of low Young's modulus directly to a flexible substrate. The second method is to assemble the device using a conductor that is itself stretchable. It is usually prepared by mixing a conductive substance into an elastic matrix.[44] Someya et al.[45] prepared a stretchable organic light-emitting diode active matrix. Printable elastic conductors, such as single-walled carbon nanotubes, are uniformly dispersed in the high elastic film of the fluorine-containing copolymer. Elongated carbon nanotubes fabricated by the ionic liquid method exhibit up to 100% elongation and $100 \text{ S} \cdot \text{cm}^{-1}$ conductivity.

In terms of geometric patterns and device design, conductive nanomaterials can be designed into stretchable structures, such as spring, mesh, [46] etc. Rogers et al. [47] first proposed the adhesion of rigid traditional inorganic materials with excellent electrical properties to the surface of elastic substrates. Inorganic semiconductors, including electronic components and connecting circuitry, are assembled on stretchable devices. Distinctively, the tension of the high Young's modulus mechanical planar layer is negligible. [48] The complex wave structure absorbs most of the tensile strain generated during the compression-relaxation process of the substrate. This island-bridge design significantly improves the stretchability of the sensor for the first time; In this design,[49] the movable module with large rigidity is used as a floating island, and the connecting line with small rigidity is used as a pull bridge. The non-coplanar structure of the deformable link, including straight strips and serpentes, allows the sensor to undergo complex deformations, such as rotation and twisting. Recently, the self-similar fractal layer is one of the choices of the connecting line, [50]and this fractal line is beneficial to adapt to different deformations. In order to meet the practical requirements, three contact modes between the connecting wire and the soft substrate are effectively utilized, including non-bonded, partially bonded, and fully bonded. For example, the non-bonded design is free to deform and the exposure of the wiring is more sensitive to physical damage,[51] Such that it exhibits the best mechanical properties.

The micro-nano patterning of metal film is one of the core technologies of flexible electronic manufacturing. Lithography is a method used to obtain high-precision micro-nano patterns. However, there are some problems, such as high requirements for processing environment and raw materials, complex process, high cost and poor compatibility with subsequent processes. [52] The problem has greatly limited the possible industrial production. With the development of technology, inkjet printing is also widely used in micro-nano patterning process, and Ahn proposed omni-directional printing technology. The conductive electrode with telescopic performance is prepared. Sun et al. [53] Proposed near-field electrospinning (NFES) to deposit solid nanofibers in a direct, continuous, and controllable manner. North Carolina State University Dong et al. achieved serpentine electrode electrophysiology based on inkjet printing by controlling the concentration of Ag NWs conductor, ink viscosity, printing speed and other parameters. Rapid preparation of the sensor. However, the inkjet printing process also has some problems, such as poor positioning accuracy, difficult ink configuration, and low efficiency of batch printing, limiting possible industrial production. Laser cutting is applied to the patterning of metal films, and the whole process can be completed within 10 minutes without any wet process. A process method with simple operation, easy parameter adjustment,

high efficiency and low cost is provided for the manufacture of stretchable electronics, can be industrially applied. [54]

The transfer of patterned thin-film structures onto elastic substrates has been widely used in the manufacture of stretchable devices. It is considered to be an important and orderly process to achieve transfer printing. [55] Fective transfer depend on that magnitude of the energy release rates of the donor and acceptor substrate, however, it is still troublesome to control the transfer process in actual operation. Methods to accomplish the transfer of a film structure to an elastomeric substrate through thermally inactivated tapes, water soluble inactivated films, etc., rely on a trained operator[56]. Personnel and specific equipment limit the development of industrial production. The adhesion of the PDMS matrix to the metal film can be adjusted by adding different proportions of curing agents to the PDMS matrix, thereby controlling the energy release rate and completing the transfer process of the film structure from the donor to the receptor. The process has the advantages of simple operation, low cost, high speed, high efficiency and good compatibility with the roll-to-roll manufacturing process, and is suitable for industrial mass production. [57]

Finally, in order to achieve the heterogeneous integration between the metal electrode and the elastic substrate, PDMS is used to encapsulate the transferred structure. A wearable flexible ECG sensor based on capacitive coupling can be prepared. [58]

3.3. Energy supply of equipment

Wearable electronic sensors require a wearable energy storage device to operate. Therefore, it is necessary to develop high safety, miniature and highly integrated energy supply and storage devices. Among them, supercapacitors, flexible batteries and self-powered systems are mainly used.

3.3.1. Supercapacitor

The wearable supercapacitor has higher power density, better rapid charge and discharge performance and better stability. Therefore, it is an ideal wearable energy supply device. Park et al. Prepared a graphene/polypyrrole/polydimethylsiloxane (PDMS/PPy/GF) composite by chemical vapor deposition and electrodeposition, Vitamin foam. [59] The foam shows excellent pressure-sensitive, temperature-sensitive and electricity storage properties, and the supercapacitor assembled based on the foam. Pressure and strain detection is a multi-functional system that can detect the pressure caused by finger touch and the strain of the skin on the back of the hand.[60]

A traditional sandwich-type supercapacitor is formed by stacking a current collector, an electrode material and a quasi-solid electrolyte layer by layer. It is not conducive to the integration and miniaturization of wearable systems, so workers are more likely to use planar interdigital supercapacitors. Shen et al. Designed a wavy carbon nanotube/polyaniline electrode. A 40% stretchable miniature supercapacitor was fabricated using lithium perchlorate gel electrolyte. A high areal capacitance ($44.13 \text{ mF} \cdot \text{cm}^{-2}$) is achieved, which allows normal operation under stretching, twisting, and winding.[61]

There are still some problems to be solved. For example, the quasi-solid electrolyte has poor stability in air and needs to be developed and optimized. In addition, the self-healing of supercapacitors also needs some research.

3.3.2. Flexible battery

Compared with flexible capacitors, flexible batteries have higher energy density and can supply power continuously for a longer time. At present, flexible wearable batteries include lithium-ion batteries, sodium-ion batteries, zinc-ion batteries and lithium-sulfur batteries. But the energy density of most batteries is much lower than expected. Due to the requirement of stretchability, in order to

prevent electrolyte leakage during deformation, there are higher requirements for battery packaging. [62] The field of flexible batteries is still at an early stage of development.

Professor Peng of Fudan University believes that due to the development of textile technology, linear batteries will be easier to achieve wearable applications. Therefore, after research, the research group proposed a new method to make parallel multi-walled carbon nanotubes into carbon nanotube fibers. [63] The silicon is deposited on the surface of the fiber by atomic layer deposition. The assembled lithium ion battery not only can utilize the high specific capacity of silicon and the excellent conductivity of the multi-walled carbon nanotube, The flexibility of carbon nanotubes can also be used to alleviate the volume change during charging and discharging, so that there will be no significant capacitance attenuation. [64]

There are still many challenges in this field, such as how to improve the safety of flexible batteries. How to reduce its mass while increasing its energy density. Therefore, it is necessary to find new materials, optimize the structure and design new electrolytes to improve the safety and functionality of flexible batteries. In addition, because the preparation of flexible batteries is slow and complex, it is also very important to develop new preparation methods. [65]

3.3.3. Other flexible functional devices

In addition to these two devices, there are other power supply devices that can be applied to wearable electronic devices, such as flexible solar cells. A friction generator, a biological power generation device, a piezoelectric device and that like.

Park et al., describe a method of using an organic photovoltaic solar cell (OPV) as an energy supply component, At the same time, it is integrated with organic electrochemical transistors (OECTs) on the parylene film, and the ZnO structure is introduced into the OPV cell. The power conversion efficiency of the battery reaches 10.5%. And that OPV has high flexibility and can be stuck on the surface of the human body to realize the detection of the human body signal. [66]

The conversion of mechanical energy into electrical energy by triboelectricity and piezoelectric principle is also sought after by many scientists. The invention has the advantages of simple structure, light weight, small volume and high energy density. Recently, Guo et al. Made silk fiber and polyvinylidene fluoride (PVDF) by electrospinning technology. [67] The fibers are deposited on the conductive fabric. Due to the large specific surface area and aspect ratio of the fibers (figs. 6h-i), the fibers are endowed with a triboelectric-piezoelectric nanogenerator. Based on the excellent stability of TPNG, the piezoelectric device can achieve a maximum power density output of $310 \mu\text{W} \cdot \text{cm}^2$. By fixing the piezoelectric device on the elbow of the subject, the monitoring of different bending degrees of the elbow can be realized. [68]

3.4. Introduction of specific preparation method

In order to better realize the initial design and test verification of wearable flexible electronics, Zhang, Professor Qiu and others proposed a rapid preparation method of flexible electronics based on "cutting and pasting". [69]

Firstly, a micro-nano patterning process based on laser cutting is proposed by comparing the photolithography process with the inkjet printing process; Then, the energy release rate was controlled by adjusting the adhesion force of the polydimethylsiloxane (PDMS) matrix. Transferring the patterned film structure to an elastomeric substrate; Then, in order to ensure the close contact between the metal electrode and the flexible substrate, PDMS was used to encapsulate the whole structure; [70] Finally, a multi-channel physiological signal acquisition system is built to test the flexible electrodes and explore the medical treatment. [71] The experimental results show that the proposed process has higher efficiency and lower cost compared with the traditional flexible

electronic processing technology. The whole process can be completed within 10 minutes, and the prepared electronic sensor can conformally contact with the skin and output stable signals. It can lay a foundation for the initial design and subsequent industrial application of flexible electronics. It can be seen that the preparation method is very fast, has high efficiency and low cost, and has great advantages.[72]

4. Summary and outlook

With the popularity of smart terminals, wearable electronic devices show a huge market prospect. It can be seen that the research on wearable flexible electronic devices has reached a deeper level. There are many theories or specific methods of preparation and use that have been discovered or invented.[73] With the further research and the extensive intersection of various disciplines, More flexible materials with good electrical properties will be developed for wearable sensors. In recent years, the printing manufacturing technology, represented by printing electronics and 3D printing, has been developed with its material universality and flexibility. Base material applicability, convenient customizability, high efficiency of large-scale continuous production, especially the cleanliness and environmental protection of its additive manufacturing process.[73] Quickly received extensive attention from scientific research and industrial production. Especially with the rapid development of printing and electronic technology, printing and printing technology, with its efficient and environmentally friendly process of adding materials, has been widely used.[74] It is promoting the emerging printing electronics industry to develop in the direction of functional and device-oriented printing manufacturing. New printed electronic devices, represented by flexible electronic devices, have shown great vitality in advanced manufacturing.[75]

However, there are still some difficulties to be overcome in this field. For example, large-scale pressure sensor arrays are very important for the development of wearable sensors in the future. Pressure sensors based on piezoresistive and capacitive signal mechanisms have signal crosstalk, which leads to inaccurate measurement. In terms of fabrication process, the current microfabrication technology represented by lithography has high processing cost and complex process. And the huge waste of materials caused by the process and the like,[76] It can not adapt to the large-scale, low-cost, efficient and clean manufacturing of high-performance flexible wearable devices in the future. In the future, this technology may play a greater role in scientific exploration, military operations and other important occasions.[77]

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