Stretchable Electrodes for Wearable Electronics

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Abstract—Nowadays, stretchable electrodes for wearable electronic devices are becoming more and more popular in human-computer interaction, communication equipment, robot, and other fields, and related research is also progressing. This paper aims to study its three fundamental technical principles: stretchable electrode materials, stretchable electrode structures, and high-adhesion substrates. This paper will first introduce the stretchable material, the physical basis for the realization of the stretchable electrode. Then we introduce the stretchable structure to explain why we choose such a structure. It also tells some possible innovative ideas on the structure. Finally, the high-adhesion substrate is introduced. The stretchable electrode also needs a carrier to maintain its structure. This paper will greatly value stretchable electrodes for wearable electronics research and application. Let's move on to the introduction section.

1 INTRODUCTION

Since its components can be compressed, twisted, and made to conform to intricate non-planar surfaces, flexible and stretchy electronics, a recently created technology, is gaining popularity and finding usage in many industries. Stretchable electronic devices and associated manufacturing technologies are now developing quickly due to wearable electronics applications that have a favorable impact on many parts of everyday life. Next-generation wearable electronic applications are made possible by flexible, soft, and stretchy electronics, opening up various applications for health care, energy, and military uses [1]. For instance, flexible transparent electrodes (FTEs), which play a crucial part in flexible electronics, have garnered significant interest in recent years. Doped metal-oxide films have dominated optoelectronics for many years. Indium tin oxide (ITO) films are an example of a doped metal-oxide film that is both optically transparent and electrically conducting. ITO films are fragile, nevertheless, and are likely to break when put under slight tension. FTEs on elastomeric substrates frequently distort with bending. Other deformation modes include folding, twisting, and stretching; stretching is the most challenging and demanding because of the significant applied forces accompanying the deformation process. Highly stretchy FTEs can also tolerate bending, folding, and twisting [2]. Such flexible electronics lead to the outburst of a new branch of study: stretchable electrodes. In particular, wearable sensors can be installed on clothing or even directly mounted on a person's skin to monitor physical signals for the diagnosis of diseases and the monitoring of one's health. Conductive electrodes have evolved into a critical component of wearable sensors and are now a standard component of electronic products. The rigidity and brittleness of conventional electrodes made of single-crystal silicon, polycrystalline metals, or metal oxide coatings restrict their use in applications requiring significant deformation or close integration with curved surfaces [3].

Highly stretchable electrodes are attracting increasing attention. This article will mainly focus on stretchable electrodes for wearable electronics. We will figure out some stretching strategies which can be used to improve the application of stretchable electrodes. This article will first introduce stretchable materials. Nanomaterials play an indispensable role in wearable electrodes. Nanomaterials can provide many new approaches for high-tech devices and manufacturing processes in various applications. The conductive material is based on four kinds of tactile sensing, and the conductive electrode is the key component to realize the sensing function. In this paper, we discuss the tensile electrode materials related to nanomaterials and analyze the advantages and disadvantages of each material.

Next, we will take a look at stretchable construction. Most materials used to make highly conductive transparent electrodes are difficult to stretch. However, by changing the structure of the materials, it is possible to increase their stretching ability on a large scale. The top-down approach creates the notched pattern on the nanocomposite sheets, drawing inspiration from the Japanese paper-cutting craft known as kirigami. The process can be scaled down to the nanoscale using a standard method like photolithography.

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Besides, Existing stretchable transparent electrodes frequently experience a significant increase in sheet resistance when stretched to high strains or repeatedly to relatively low stresses over thousands of cycles. Strain fatigue may be removed entirely, and tension can be significantly increased by tuning the topological structure of metal nanoparticles and altering adhesion. All these things could be ideal electrodes for flexible electronics.

Last, we will discuss chemical bonding in highly stretchable and transparent electrodes. The scientists aimed to build a solid covalent link (gold-sulfide bond) between a gold nanomesh electrode and the underlying elastomeric substrate to boost the adhesion since the contact between the metal and the substrate is done with der Waals forces. Since tracking human motion is crucial and human motion analysis is also required, e-textile technology has a bright future because of its low cost, lightweight, flexibility, and other advantages. It is also one of the upcoming uses for developing high-adhesion substrates.

The research of this paper will be of great value to the research and application of stretchable electrodes. We hope that these studies can effectively help with wearable electronic devices.

2 STRETCHABLE MATERIALS

As we mentioned in the abstract, nanomaterials play an irreplaceable role in wearable electrodes. Nanomaterials can provide many new methods for high-performance devices and manufacturing processes in various applications. Conductive materials are based on the working principle of four types of tactile sensing, and the electrode with conductivity is the key component to realize the sensing function.

Since most fiber materials are insulators (such as natural cotton fibers and artificial polyester fibers), it is necessary to attach functional conductive materials on flexible fiber substrates by unique preparation methods to obtain fiber electrodes with conductive properties.

At present, common conductive materials include conductive metal materials (such as silver, copper, etc.), carbon conductive materials (such as graphite, carbon black, carbon nanotubes, graphene, etc.), metal compound materials (such as two-dimensional titanium carbide) and conductive polymer materials (such as polyacetylene, polyaniline, polypyrrole, polythiophene, and its derivatives) [4].

Tensile electrode materials related to nanomaterials include transparent conductor electrodes, organic light emitting diode, Thermoelectric element sensors, film solar cells, etc.

Indium tin oxide films are most commonly used in optoelectronic devices in the current transparent conductor industry. For example, we use it on thin film solar cells, flat panel displays, and touch screen panels. However, the supply of its key raw material, indium is unstable, the productivity is low, and the market price has been rising.

Carbon-based nanomaterials (carbon nanotubes and graphene), noble metal (gold, silver) nanowires, and microgrids have been widely used to manufacture transparent conductors. However, carbon-based nanomaterials usually cannot meet the requirements for efficient, transparent conductors in many cases. Precious metals (gold and silver) exhibit excellent conductivity and transmissivity. However, because they are too expensive as raw materials, they lack competitiveness in a large area and low-cost transparent conductors.

The practical use of copper nanowires as a transparent conductor is limited, mainly because it is easy to oxidize at high temperatures. In our intuition, copper is not an easily oxidized metal. However, we may have overlooked that copper with nanostructures has a larger specific surface area than bulk copper. The contact area with air is expanded, making it easier to oxidize.

Flexible transparent conductors and stretchable electrodes based on copper nanowires percolation network are reported. Its manufacture involves ultra-fast plasma nanoscale welding using circularly polarized lasers under environmental conditions, and room temperature is used to minimize copper oxidation problems.

To successfully produce this power supply, we can consider using liquid metal as the electrode. Someone proposed a liquid metal-based triboelectric nanogenerator (LM-TENG), which uses Galinstan as the electrode and silicone rubber as the triboelectric and packaging layer. Young's modulus of liquid metal is small, and the tensile capacity is strong, enabling the electrode to maintain continuous conductivity under deformation (stretching to 300% strain). The surface oxide layer of Galinstan is dense, which effectively prevents the liquid Galinstan electrode from further oxidation and penetration into the silicone rubber. No additional sealing is required, so it has good device stability.

Operates in 3 Hz single electrode mode with an area of $6 \times 3\ cm^2$, generates 354.5 V open circuit voltage, 123.2 nC transferred short circuit charge short circuit current of 15.6 μA and the average power density of 8.43 mW/m^2 are outstanding performance values of TENG. In addition, LM-TENG maintains stable performance under various deformations, such as stretching, folding, and twisting. Different forms of LM TENG, such as block, bracelet, and fabric, can obtain mechanical energy from human walking, arm swinging, or hand clapping to drive wearable electronic devices [5] continuously.

The mechanical energy generated when the human body constantly moves is a potential power source. We have always thought of converting this mechanical energy into the power of wearable devices. To be honest, this idea is very challenging, but we are moving towards this goal.

TENG has the natural advantages of low density, simple composition, high integration, and low cost. In addition, TENG can also obtain energy from irregular and low-energy human movements. Therefore, we believe there is a feasible manufacturing strategy for wearable electronic devices, which is to use TENG material.

3 STRETCHABLE STRUCTURE

Most materials that make transparent electrodes with high conductivity are hard to stretch. However, by changing the structure of the materials, the ultimate strain capacities can be increased on a large scale.

Inspired by the Japanese paper-cutting art kirigami, the top-down technique is applied to the nanocomposite sheets to make the notched pattern [6]. The process can be developed to the microscale using a standard method like photolithography. Before failing, the nonpatterned material revealed an ultimate strain of 4%, the predominant deformation comprised stretching individual cellulose fibers at the nano, micro, and macro scales inside the matrix. However, the ultimate strain of the nanocomposite sheet can be increased to 370% after the regular notched pattern is applied to the sheet.

To investigate the impact of the paper-cut pattern on mechanical behavior, beam deflection analysis is used. It shows rigid materials with a kirigami pattern display high flexibility and can withstand high stress as the stress distribute to the present deformation points. Finite-element modeling can accurately predict the patterned nanocomposite's tensile behavior and geometric characteristics. The notch's vertical and horizontal spacing and length can vary, influencing the materials' critical buckling load and extensibility. The material is weakened, its buckling load is lower, and extensibility is improved through increasing notch length. However, notch spacing increases the sheet's rigidity and assigns a higher buckling load. The variations of the buckling load caused by the material's deformation are underestimated by the finite element analysis prediction, particularly around the cut's vertex. The finite element model does not represent the ripping and fracture in these regions.

Nanocomposite sheets with pattern defects have a high degree of damage tolerance. The applied force of the material is evenly distributed throughout the piece rather than concentrated in a few random starting points. Three-dimensional paper-cut nanocomposites have high conductivity and high resistivity. This property can be used for argon-filled glass tube tunable plasma discharge electrodes. Throughout the strain regime, the electrical conductivity of stretchable nanocomposite remains constant. As the strain level increases, a higher degree of local ionization and plasma intensity is due to the increased recombination of ionized material. The development of tunable electrodes opens up the possibility of many practical new applications, such as stretchable-electronic technology [6].

Failure occurs at low strain levels of cyclic loading due to fatigue of strained materials. However, metallic materials frequently display high cycle fatigue, which is always a drawback. When highly stretched or repeatedly stretched to relatively modest stresses for thousands of cycles, the sheet resistance of existing stretchable transparent electrodes often increases considerably. By optimizing the topological structure of metal nanomaterials and modifying adhesion, strain fatigue can be eliminated, and tension can be significantly enhanced [7].

First, the gold nanomeshes are well connected, and the nodes on the gold nanomesh play an essential role in restoring the original shape of the metal nanomesh after removing the stress. Pre-strained gold nanomeshes with optimizing topology lose a few percent of transmittance but gain a considerable stretchability. When stretched to 300% strain, gold nanomesh maintains low thin layer resistance and excellent transparency and shows no fatigue after 50,000 stretches to up to 150% strain.

Second, ligaments in a gold nanomesh on a smooth substrate can be relocated and redirected locally during stretching to relax and return to their original configuration when the stress is removed. The conductor demonstrates excellent fatigue resistance by changing the adhesion between the gold Nanonet and the underlying substrate.

Moreover, cells can grow on gold nanomesh as they are porous and conductive to biological macromolecules in fluids. Conductive, biocompatible, and completely fatigue-free metal meshes will be ideal electrodes for both flexible and implantable electronics [7].

4 HIGH ADHESION SUBSTRATE

In recent years, ITO has dominated the broadband field. It is a common material for electrodes in electronic materials such as liquid crystal displays, touch screens, and solar cells. ITO has a microstructure of (In_2O_3:SnO_2=9:1) by doping In_2O_3 with Sn, and under oxygen-deficient conditions to form a carrier concentration of 1020 to 1021 cm^{-3} and a mobility of 10 to 30 cm^2/vs. so ITO has the conductivity of a semiconductor. Because ITO is a broadband thin film material with a band gap of 3.5-4.3 eV. The ultra-violet light region produces an absorption threshold of 3.75 eV, so the penetration rate of the ultra-violet light region for ITO films is meager. Hence, ITO has good electrical conductivity and high light transmission in the visible area. In recent years, since ITO films are too brittle and indium is scarce, some scientists have tried to find a substitute for ITO. In the previous ten years, solution-processed metal nanowire networks, carbon nanotube networks, graphene sheets, and templated metal nano-interconnects have received attention and some research results. Even though these have been comparable to ITO films, these new electrodes cannot be widely used because of the poor adhesion of conductive materials and flexible bases.

As a result of the metals and the substrate's interaction with Van der Waals forces, the scientists tried to introduce a robust covalent bond (gold-sulfide bond) instead of a der Waals force between the underlying elastomeric substrate and the gold nanomesh electrode to enhance the adhesion. By injecting gold-sulfide adhesives, gold nanomesh can firmly adhere to the substrate and withstand several megapascals' pressures. By successfully improving stretchability through pre-training methods, the material resistance does not change much in a single stretch of 25,000 cycles of periods of 50% or 160%, making this conductor the perfect foundation for powerful stretchability. Improving scratch resistance by introducing covalent bonds can also be applied to many systems, such as Ag nanowires and carbon nanotube films [8].

On the other hand, human motion analysis is also necessary. It is essential to track human motion, which is helpful in many fields, so e-textile technology has a promising future due to its low cost, lightweight, flexibility, etc. It is one of the future applications for developing high-adhesion substrates. Modern textiles are usually classified into three categories, fibers, yarns, and fabrics, fiber-based strain sensors can be obtained from strain-sensing materials, and fabric-based sensors are developed directly from sensing fibers or fabrics coated with strain-sensing materials. Scientists can determine human activity by monitoring the strain in the threads. For example, using graphene-woven fabrics, it is possible to detect facial expression changes, breathing, blinking, and pulses very well.

Similarly, by installing sensors at the throat and collar, it is possible to recognize the pronunciation of different words accurately. With the help of these multifunctional sensors, an early warning system for children and people with breathing difficulties can be effectively established. However, there are also some difficulties in detecting the anatomical horizontal plane and around the principal inertial axis. Because of the small radius of rotation, the detection results will have significant errors, so this problem may be one of the main directions to be overcome in the future. At the same time, the manufacture of textile sensors with better flexibility and sensitivity and the establishment of effective links with other units, such as power, signal processing, etc., is also the main future development direction [9].

5 DISCUSSION

This article discussed several aspects of stretchable electrodes, including the material, stretchable structure, and high-adhesion substrate.

Several nanomaterials are suitable for making highly conductive, flexible, and transparent electrodes. For example, ITO film has good electrical conductivity and is optically transparent. In recent years, it's commonly used in optoelectronics, such as touch screen panels and solar cells. However, IOT films are very brittle and easy to break when applying a minor strain [1]. Also, the productivity of its raw material, indium, is low, and its price is rising. The other type of material is a liquid metal called LM-TENG. LM-TENG performs high stability under the strain of 300%. It also contains the natural benefits of lightweight, small size, and inexpensive materials. However, it still has various problems that need to be solved, like the controlled motion of LM-TENG [1].

Through structural design, the nanomaterials can perform high deformability regardless of nanomaterials' size or mechanical properties. Kirigami notched pattern gives high stretchability of the nanomaterials. The ultimate train of nanocomposite sheets with a top-down technique is up to 370%. The materials with kirigami patterns can be stretched to micro/macroscales. In addition, the electrical conductivity of patterned sheets remains constant throughout the strain range. Kirigami is applied to various fields, such as skin-like electronics and bioinspired soft robotics [2].

Gold nanomesh with optimized topological structure and modified adhesion can minimize strain fatigue. The cracks can distribute evenly among the whole metal sheet. It still maintains a high electrical conductivity and stretchability. However, proper adhesion is essential to the flexible behavior of the gold nanomesh. Both weak and robust adhesion will affect the stretchability [2].

The solid covalent bond replaces the Van der Waals forces between the metal and substrate to improve the adhesion of conductive metals and stretchable substrate. As a result, the nanomesh adhered firmly to the substrate, and gold nanomesh could bear high pressure. The material resistance won't decrease a lot through thousands of stretch cycles.

6 CONCLUSION

This article focuses on developing technology with stretchable electrodes and various sensors because these developments can undoubtedly retreat many fields. ITO films are certainly one of the most mainstream electrodes today. We discuss the shortcomings of ITO films and how they can be improved, for example, by applying wearable sensors or adding chemical bonds in the material field. We need to strengthen ITO films because they are fragile and may break when subjected to slight tension and because the supply of indium, a critical raw material, is unstable, productivity is low, and market prices have been rising, making the industry unable to rely on ITO films for long. Currently, gold nanomesh technology is well developed, firstly, the gold nanomesh is well connected, and when stretched to 300% strain, the gold nanomesh maintains low resistance and good transparency and remains reliable after 50,000 stretches after pulling to 150% strain. By developing stretchable electrodes for wearable electronic devices, we can solve problems in areas such as medical and military, and there is still room for technological improvement.

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REFERENCES

1. W. Wu, "Stretchable electronics: functional materials, fabrication strategies and applications," Sci. Technol. Adv. Mater., vol. 20, no. 1, pp. 187–224, Dec. 2019.
2. S. Huang, Y. Liu, Y. Zhao, Z. Ren, and C. F. Guo, "Flexible Electronics: Stretchable Electrodes and Their Future," Adv. Funct. Mater., vol. 29, no. 6, p. 1805924, Feb. 2019.
3. S. Zhao et al., "Recent advancements in flexible and stretchable electrodes for electromechanical sensors: strategies, materials, and features," ACS Appl. Mater. Interfaces, vol. 9, no. 14, pp. 12147–12164, Apr. 2017.
4. S. Han et al., "Fast plasmonic laser nanowelding for a cu-nanowire percolation network for flexible transparent conductors and stretchable electronics," Adv. Mater., vol. 26, no. 33, pp. 5808–5814, Sep. 2014.
5. Y. Yang et al., "Liquid-metal-based super-stretchable and structure-designable triboelectric nanogenerator for wearable electronics," ACS Nano, vol. 12, no. 2, pp. 2027–2034, Feb. 2018.

6. T. C. Shyu et al., “A kirigami approach to engineering elasticity in nanocomposites through patterned defects,” Nat. Mater, vol. 14, no. 8, pp. 785–789, Aug. 2015.
7. C. F. Guo et al., “Fatigue-free, superstretchable, transparent, and biocompatible metal electrodes,” Proc. Natl. Acad. Sci. U.S.A., vol. 112, no. 40, pp. 12332–12337, Oct. 2015.
8. S. Huang, Y. Liu, C. F. Guo, and Z. Ren, “A highly stretchable and fatigue-free transparent electrode based on an in-plane buckled au nanotrough network”, Adv. Electron. Mater., vol. 3, no. 3, p. 1600534, Mar. 2017.
9. C. F. Guo, Y. Chen, L. Tang, F. Wang, and Z. Ren, “Enhancing the scratch resistance by introducing chemical bonding in highly stretchable and transparent electrodes,” Nano Lett., vol. 16, no. 1, pp. 594–600, Jan. 2016.