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Transparent electrodes in neuroscience: Review

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Abstract:

Transparent electrodes have received increasing attention in neuroscience area such as neural interfaces, optogenetics and implantable neural sensors due to their unique ability to combine electrical conductivity with optical transparency, enabling simultaneous neural recording and stimulation without hindering optical techniques. This review concludes transparent electrodes into five categories including metal oxide-based, metal nanowire-based, conductive polymer-based, carbon-based as well as Hybrid materials and composite structure. Starting from the most widely used and earliest transparent material ITO, this review progressively covers the latest advances in hybrid materials and composite structures. For each classical material in different categories, their drawbacks, merits and current situations are introduced. Then the applications of transparent electrodes in the field of neuroscience are introduced, especially in relation to specific disease treatments. The application prospects of transparent electrodes in the field of neurology are broad, but they still face many technical challenges and problems. The future research should explore long-term implanted neural interfaces.

Keywords: Transparent electrode; neuroscience; hybrid material.

1. Introduction

Neurology is the study of the nervous system, which is responsible for transmitting sensations, controlling movement, regulating internal organs, and processing emotions. In the study of neuroscience, the response of the nervous system to disease is often studied. In the process of understanding this phenomenon, electrode technology has become a key tool in the field of neuroscience. Traditional metal electrodes have certain limitations in neurological applications such as neural interfaces and optogenetics. Among them, neural interfaces or optogenetics applications usually require simultaneous electrophysiological recording and optical imaging. Traditional metal electrodes, however, obstruct the optical signal pathway during this procedure. Simultaneously, they may also produce artifacts when the laser is irradiated on the metal electrodes, which interferes with the accuracy of electrophysiological data [1-3]. Transparent electrode materials have effectively addressed these issues.

Transparent electrodes refer to materials that have high conductivity and high transmittance in the visible light range, which allow efficient propagation of light while maintaining good conductivity, and can record high-resolution electrophysiological signals. Therefore, the application of transparent electrodes can associate image data with neural signals, that is, light stimulation can be directly transmitted to the part of the brain where the electrode obtains data [1,4,5]. Moreover, transparent neural electrodes also have excellent flexibility and biocompatibility. Transparent electrodes can be traced back to the 20th century, when metal-based transparent electrodes such as ITO were first used in optoelectronic devices. Nonetheless, with the higher performance requirements of transparent electrodes, different transparent electrodes have emerged, such as metal nanowire-based electrodes, conductive polymer-based electrodes, and carbon-based electrodes. Subsequent to 2000, these novel electrode materials have garnered significant interest and have become pivotal in the domain of neurology. With the further development of materials science, hybrid materials and composite structure electrodes have emerged, and their performance has made them widely mentioned and used in the field of neuroscience.

This review aims to classify the existing transparent electrode materials into five categories according to the material category and summarize them. In the meanwhile,it explains the different materials and their synthesis methods, advantages and disadvantages, and their applications in the neuroscience field. In addition, this review explores the application of transparent electrodes in the neuroscience field and their future development, and summarize them.

2. Types of transparent electrode materials

Transparent electrodes are crucial in optoelectronic applications, especially in neurology. To adapt to different application environments, transparent electrodes made of different materials have been developed. To facilitate in-depth research by researchers, transparent electrodes are divided into five kinds according to material categories: metal oxide-based electrodes, metal nanowire-based electrodes, carbon-based electrodes, conductive polymer-based electrodes as well as hybrid materials and composite structure electrodes.

2.1 Metal oxide-based transparent electrode materials

Metal oxide-based electrodes, also known as transparent conductive oxides electrodes (TCOs electrodes), are the earliest and most widely used transparent electrode materials. TCOs are metal oxide-based materials that are both conductive and have relatively low electromagnetic wave absorption rates in the visible light range, that is, they do not absorb a large amount of visible light and allow light to pass through. The combination of oxygen and different metal elements constitutes composite semiconductor TCOs, whose photoelectric characteristics can be adjusted by selecting different metals or adding dopants. The SEM cross-sectional image of some specific TOCs is depicted in Figure 1 a, which also shows the planar SEM of them. Common TCOs with excellent optical characteristics include tin, indium and zinc oxides, etc. Next, this section will list several classic metal oxide-based transparent electrode materials used in the field of neuroscience [6].

ITO:

ITO is a widely used transparent metal oxide-based material. Due to its good electrical conductivity and light transmittance, it is widely used in the field of neurology, especially in biosensors. Through thin film techniques such as sputtering deposition and electron beam evaporation, ITO is uniformly deposited on a specific base to form a very thin conductive transparent layer. This transparent layer is called ITO film and is extremely suitable for use as a transparent electrode for electronic devices. Due to the scarcity and high consumption of indium resources worldwide, as well as the limitations of deposition technology, the cost of ITO does not show an advantage, and the mechanical properties of ITO are also relatively poor. In order to overcome these difficulties, other alternative materials have been produced [7,8].

ZnO:

ZnO is a metal-based material with a wide bandgap made of zinc metal, which means it absorbs little visible light and is transparent. Zinc is an abundant metal with a sufficient supply and a stable price. Compared with ITO, the cost of ZnO is significantly lower [8]. As an alternative material to ITO, ZnO solves the problem of high cost, but it still has poor mechanical properties and will break under high stress conditions [9]. To solve this problem, different materials are doped into ZnO to create novel materials to solve the brittleness problem.

AZO:

AZO is a ZnO-based doping material, which is essentially a variant of ZnO, formed by doping aluminum into ZnO. Due to the rich aluminum and zinc metals, AZO is also lower in cost than ITO. The doping of aluminum metal makes the material have better conductivity. The appropriate amount of doping metal in ZnO can improve the conductivity of ZnO film, among which the doping of aluminum metal is effective. At the same time, AZO can also be obtained through thin film preparation methods such as magnetron sputtering, spray pyrolysis, pulsed laser depo-

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sition process, vacuum evaporation, etc. to obtain high transmittance and high conductivity films [10]. Research shows that the elastic recovery of AZO film is 37.5%, and the critical fracture load is about 91mN, which shows that AZO has good adhesion and mechanical durability [11].

Other metal oxide-based electrodes, such as titanium oxide TiO2, are neurotoxic when in the form of neural particles, such as oxidative stress and inflammatory response. Therefore, TiO2 is not suitable for use in the neurological field [12]. Another example is FTO, which is more used in photovoltaics and real-world technologies and has not yet been widely studied in the neurological field. It may be applied to the neurological field in the future. There are still many such metal oxide-based light-transmitting electrode materials that are not suitable for use in the neurological field. This section will not be described in detail, and the same applies to the subsequent sections.

2.2 Transparent metal nanowire-based electrodes

Traditional metal-based electrodes have extremely high conductivity, but they are usually not light-transmissive because traditional metals have extremely high light absorption. In order to address the limitations of traditional metal-based electrodes, researchers have developed transparent metal nanowire-based electrodes (MNWs),whose cross-sectional SEM image is depicts in Figure 1b and 1c. Metal nanowire electrodes composed of transparent, flexible and highly conductive networks formed by interweaving metal nanowires including but not limited to gold nanowires, silver nanowires, copper nanowires, etc. are called transparent metal nanowire-based electrodes.

Typically, as the diameter of MNWs increases, light transmittance decreases, whereas longer wire lengths improve conductivity. To produce high-performance MNWs with consistent morphology and high yields, a range of synthesis techniques have been developed, with the most prominent being template-based methods and chemical approaches. The template-based method for synthesizing MNW is widely used in the early stage and is divided into hard templates and soft templates. Metal atoms grow into nanowires along the pre-designed template material structure. This method is the hard template-based method. However, the hard template-based method easily destroys the large-scale purification process of nanomaterials and is not suitable for large-scale practical applications. In relative terms, the soft template-based method induces the growth of metal nanowires through chemical substances such as surfactants, which has the advantages of simplicity and low cost. However, the nanowires synthesized by this method have a low aspect ratio and yield, and may also have surface defects.

Compared with the template-based method with relatively low production efficiency, chemical synthesis methods are more conducive to the morphology control of MNW, including polyol method and hydrothermal method. The polyol method refers to the use of polyols to reduce inorganic salts to generate MNW under high temperature environment. This process usually requires solutions and reducing agents. This method is suitable for large-scale industrialization and is inexpensive. The hydrothermal method does not rely on any organic solution. Apart from its high-pressure conditions and the requirement for additional safety measures, this method is environmentally friendly, efficient and precise. MNW has become one of the most promising materials in the field of neuroscience because of its excellent conductivity, transparency, low cost and flexibility. The main types of metal nanowire-based electrodes include gold nanowires, silver nanowires, copper nanowires, platinum nanowires and nickel nanowires, each of which has its own advantages and disadvantages [13-15].

Gold nanowires:

Compared to traditional ITO electrodes, transparent nanomesh electrodes made of gold nanowires are more flexible and have excellent biostability, making them suitable for long-term implantable neural electrodes. However, due to the high cost of gold itself and the complex process of preparing gold nanowires, it is difficult to produce gold nanowire electrodes on a large scale at low cost [16,17].

Silver nanowires:

Nanomesh electrodes made from silver nanowires are also flexible, transparent, and conductive. Compared to transparent gold nanomesh electrodes, their preparation methods such as spraying and inkjet printing are more conducive to low-cost large-scale production. Although silver's conductivity is reduced by long-term exposure to corrosive chemicals, it can be used in long-term implants by adding a protective coating [18,19].

Copper nanowires:

Copper metal wire mesh electrodes are also used as substitutes for gold nanowire mesh electrodes. While having the same conductivity as gold, they have lower raw material costs and more abundant reserves. However, due to the disadvantage that copper will oxidize over time, some methods to prevent oxidation can be used to prevent this problem, such as surface coating protection, chemical doping, self-encapsulation technology, etc [20].

Platinum nanowires:

Platinum nanowire mesh electrodes made of platinum nanowires have the characteristics of high transparency, high flexibility, and low sheet resistance. Compared with the oxidation problem of copper metal, platinum metal has very stable chemical properties, that is, electrical stability under special conditions. This property makes platinum nanowire mesh electrodes suitable for long-term neural

implant devices [21].
Nickel nanowires:
Similarly, nickel nanowire mesh electrodes have the advantages of high transparency and high flexibility. However, the mesh electrodes composed of nickel nanowires still need surface treatment to overcome oxidation reactions. At the same time, the transparent electrode resistance is relatively high and the optical performance is poor. Therefore, compared with other metal nanowire mesh electrodes, this metal does not have the potential for some high-end applications such as brain-computer interface and optogenetics, but it still has the potential for application in flexible neural electrodes and neural sensors [22].

2.3 Conductive polymer-based transparent electrode materials

Transparent conductive films made of conductive polymers as the main material are called conductive polymer-based transparent electrode materials, and Figure 1d to Figure 1f show the SEM images of three common conductive polymer-based materials. The synthesis methods of this material are generally divided into electrochemical polymerization and oxidative chemical polymerization [23]. The former requires a conductive substrate, and the latter requires an oxidant. Conductive polymer films can be prepared by three different methods: coating treatment, polymerization solution deposition, and chemical vapor deposition. Conductive polymers combine the characteristics of metals and traditional polymers, such as high conductivity, high light transmittance, strong adhesion, and high stability [24]. The types of conductive polymer-based transparent electrode materials currently used in the field of neuroscience mainly include PEDOT, PANI, etc.
PANI:
PANI is an early conductive polymer with a research history of nearly 200 years. The polymerization mechanism of PANI has been varied, including free radical coupling mechanism, chemical synthesis mechanism, electrochemical synthesis mechanism, anion insertion mechanism, gas phase plasma polymerization, etc. Different polymerization mechanisms have resulted in various preparation methods of PANI. The properties and structure of PANI vary according to different preparation mechanisms [25]. However, PANI has poor solubility and is not suitable for solution processing to prepare thin films. And it is only light-transmissive under certain conditions.
PEDOT:
Both PEDOT and PEDOT:PSS are conductive polymers based on PEDOT, so they can be classified as the same type of transparent material. Although they are the same type of polymers, PEDOT:PSS is a modified material of the former and exhibits better solubility, flexibility and processability, so this section will focus on PEDOT:PSS. This material requires the oxidative polymerization of PEDOT in the presence of a PSS matrix. During this process, electrostatic interactions bind PEDOT and PSS together [26]. Among them, PSS provides PEDOT:PSS with the ability to form an aqueous dispersion, which means that PEDOT can be processed by conventional solutions to prepare conductive films, which greatly simplifies its application in industry.

2.4 Carbon-based transparent electrode materials

Due to the limitations of metal oxides, novel transparent electrode material substitutes are needed. Carbon-based transparent electrode materials have high conductivity and high light transmittance. At the same time, due to its abundant carbon resources and simple preparation process, it has become one of the substitutes for metal oxide-based transparent electrodes. Carbon-based transparent materials can be mainly divided into two types: carbon nanotube electrode materials whose SEM image is depicted in Figure 1g and graphene electrode materials whose SEM image is showed in Figure 1h.

2.4.1 Carbon nanotube electrode materials

The preparation of carbon nanotube electrode films is mainly divided into single-walled nanotubes and multi-walled nanotubes. Due to the high cost and low availability of single-walled nanotube preparation methods, the preparation is mostly based on multi-nanotube methods, which can also be nanotube preparation methods mainly include spraying, chemical vapor deposition, ink-jet printing, PDMS template transfer, etc. Among them, chemical vapor deposition can achieve low cost, large-scale production, high conductivity, and high light transmittance [27,28].

2.4.2 Graphene electrode material

Graphene is a two-dimensional carbon material composed of sp2 covalent bonds. Currently, the methods for preparing graphene include mechanical exfoliation, epitaxial growth, chemical vapor deposition (CVD), liquid phase exfoliation and redox method. Similar to carbon nanotube electrode materials, CVD preparation is also a more popular method for graphene preparation because of its applicability in large-scale industrial production [29].

2.5 Hybrid materials and composite structure electrodes

Hybrid materials and composite structure electrodes refer to the combination of one-dimensional carbon nanotubes and two-dimensional graphene sheets. SEM image of typical SWCNT/Gr hybrid thin film formed by traditional spin coating is presented in Figure 1i. Spray deposition and spin coating are common fabrication methods for this

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electrode, and these techniques enable the creation of uniform CNT-graphene films. This film has better mechanical properties than a single graphene or carbon nanotube, and its electrical conductivity can be further improved by doping metal particles into the film. The resulting hybrid material and composite structure electrode possesses significant electrical conductivity and mechanical flexibility while possessing high transparency [29,30].

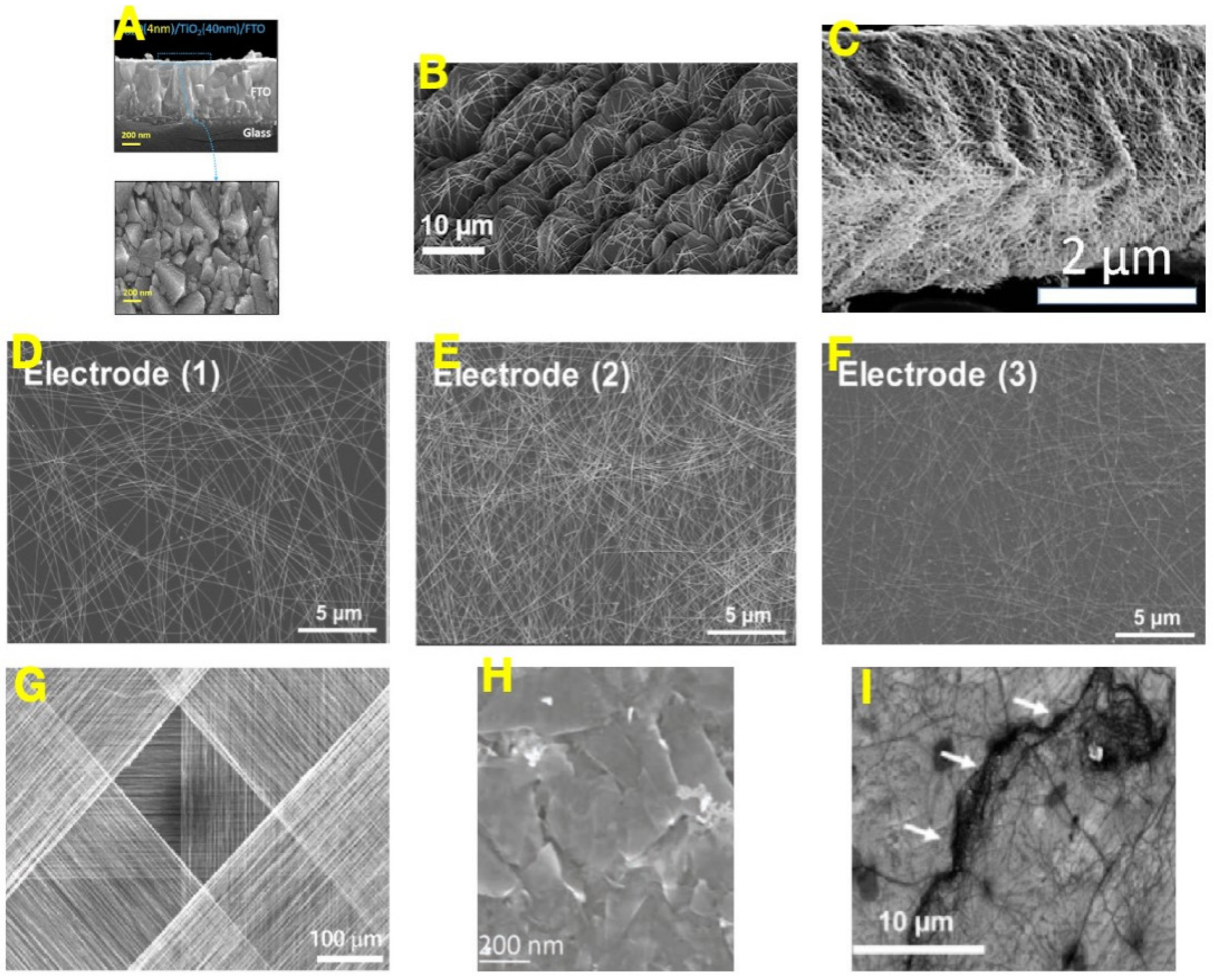

Fig. 1 Metal oxide-based transparent electrode materials. a) SEM cross-sectional image of metal oxide transparent materials such as AgxO/TiO2/FTO/Glass, with arrows indicating the corresponding planar SEM image [31]. b) SEM images of metal nanowire-based transparent materials, including AgNW coated on a textured Si surface (c) and AgNW coated on polyester/rubber wire [32]. d)~f) SEM images of three conductive polymer-based nanowire electrodes [33]. g) SEM images of carbon-based transparent materials, including four stacked nanotube thin sheets h)and the SEM image of the surface of a graphene film [34]. i) SEM image of composite materials, such as the typical SWCNT/Gr hybrid thin film formed by traditional spin coating [30].

3. Application of transparent electrodes in neural devices

Transparent electrodes have been widely used in the field of neurology, such as neural recording, neural stimulation, optogenetics, brain-computer interfaces, and flexible devices. In neural recording, transparent electrodes allow researchers to directly observe neuronal activity and record electrophysiological information in the brain at the same time. Transparent electrodes can simultaneously perform high-resolution electrophysiological recording and optical imaging, such as neural micromotor arrays. In neural stimulation, transparent electrodes can allow the interaction between light and neurons and the transmission of electrical signals, such as deep brain stimulation and peripheral nerve stimulation. In optogenetics, transparent electrodes can be used to study brain function or treat neurological diseases. In addition, transparent electrodes can also be used for prosthetic control and the development of neurological disease rehabilitation equipment through brain-computer interfaces. Finally, flexible devices, including long-term implantable devices, can also monitor long-term neural activity.

In specific, especially in diseases treatment area, some researches prove the applications. In 2017, researchers achieved simultaneous neural electrical stimulation and in vivo monitoring in mice through transparent graphene electrodes, which can enable researchers to better understand the brain circuit patterns of Parkinson's patients [35]. The DBS used to treat Parkinson's patients has a similar working principle to the above-mentioned experiment, that is, electrodes are implanted in the brain for a long time to apply electrical stimulation to specific areas of the brain, thereby alleviating Parkinson's [36]. In the same year, a portable wireless electrocorticogram (ECoG) system for the treatment of epilepsy was verified in rats. The system can record brain activity and perform electrical stimulation therapy. Through wireless transmission, ECoG can transmit brain electrical activity signals to mobile phones and analyze epileptic seizures through cloud processing. This experiment demonstrated the potential of this system in the treatment of neurological diseases. This system provides a new method for epilepsy treatment [37]. In 2018, the experiment used rats as a model to study the relationship between neural activity and emotional regulation by stimulating or inhibiting specific areas of their brains. The researchers observed behavioral changes in rats and the release of neurotransmitters in their brains to understand the neural mechanisms and causes of depression [38]. In 2022, the experiment used optogenetics to intervene in neural activity in specific brain regions and precisely regulate neuronal activity through light stimulation, as well as how to alleviate Alzheimer's disease by combining non-pharmacological techniques [39].

4. Conclusion

The key role of transparent electrode materials in the field of neurology is reflected in the simultaneous transmission of photoelectric signals. Because of the advantages of transparent materials such as flexibility, biocompatibility, low invasiveness and high transparency, transparent electrode materials are particularly suitable for neurology fields such as optogenetics, neural interfaces and brain-computer interfaces. These advantages enable the materials to play an important role in applications such as high-precision neural signal recording, stimulation, and repair.

The application prospects of transparent electrodes in the field of neurology are broad, but they still face many technical challenges and problems. For example, the application of long-term implanted neural interfaces. Highly flexible, self-repairable and biodegradable materials may become the focus of future research to reduce damage to neural tissue and rejection reactions. Another example is the problem of large-scale production. Graphene electrode materials have excellent performance, but are still limited to immature large-scale production processes. The future development of transparent electrodes in the field of neurology has great potential. Through in-depth research and combination of different fields of materials science, engineering, and neurology, transparent electrodes are expected to become a key technology for the treatment of diseases in the field of neurology.

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