Carbon-Based Textile Sensors for Different Physiological-Signal Monitoring

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As the focus on physical health increases, the market demand for flexible wearable sensors increases. Textiles combined with sensitive materials and electronic circuits can form flexible, breathable high-performance sensors for physiological-signal monitoring. Carbon-based materials such as graphene, carbon nanotubes (CNTs), and carbon black (CB) have been widely utilized in the development of flexible wearable sensors due to their high electrical conductivity, low toxicity, low mass density, and easy functionalization.

carbon-based materials   textile sensors   physiological-signal monitoring

1. Introduction

In the field of flexible wearable sensors for human physiological-signal monitoring, the main sensors involved include ECG electrodes/sensors, sensors for body motion monitoring, body temperature sensors, body humidity sensors, tactile sensors, etc. [1]. To stick to the human body for long-term use, these textile sensors need to be lightweight, breathable, flexible, stretchable, and washable [2][3]. There has been a lot of research on these textile sensors for monitoring different physiological signals.

2. Carbon-Based ECG Textile Electrodes/Sensors

Cardiovascular disease has become the highest cause of death with the advent of the aging society. Long-term ECG signals can help identify and treat potential cardiovascular disease risks. Therefore, it is important to develop ECG electrodes/sensors with high performance and long-term application [4]. Conventional Ag/AgCl ECG electrodes, also known as “wet” electrodes, use a gel to reduce their resistance to contact with human skin. However, in some cases, skin irritation and allergic reactions have been reported due to the long-term use of gel “wet” electrodes. In addition, Ag/AgCl electrode performance declines over time due to the drying of the gel [5]. Therefore, conventional Ag/AgCl electrodes are not conducive to long-term monitoring applications. One solution is to make dry electrodes from conductive textiles. These electrodes are highly comfortable and suitable for long-term monitoring applications [6].

2.1. Graphene-Based ECG Textile Electrodes/Sensors

Graphene with high conductivity is used in the study of ECG textile electrodes and sensors. In 2015, Yapici et al. proposed a GO-wrapped textile electrode for ECG monitoring. The first step was to prepare GO suspension. A total of 2 g of graphite powder (particle size less than 20 μm) and 7 g of potassium permanganate was added into 50 mL concentrated sulfuric acid, and then distilled water was added and stirred in. GO was stripped off through centrifugation and sonication. The GO suspension was obtained by hydrochloric acid treatment and distilled water washing. Finally, the nylon textile with a hydrophilic surface was immersed in it and then placed on a hydrophobic carrier. After drying, the conductive nylon was evenly wrapped with graphene. The GO textile electrode was prepared. Before and after coating GO, the conductivity of nylon changed from 6×10^{-12} S/cm to 4.5 S/cm. There was a huge increase in conductivity. Nylon has a higher conductivity when coated with GO because of its low surface roughness [7]. Compared to conventional Ag/AgCl electrodes, the GO electrode shows better performance in ECG measurements with higher fidelity to the signal. In addition, it even could be used for EMG or EEG monitoring, as in the following study [8]. In 2017, Yapici et al. improved the performance based on the above and investigated possible applications of dry electrodes. A piece of nylon textile was immersed in a GO suspension and heated to form a conformal coating of GO sheets on the textile. Finally immersed in hydrogen iodide (HI), the GO was reduced and converted to graphene to form a highly conductive graphene-coated textile. The prepared sample was cut into a small size of about 6 cm × 3 cm to form the ECG electrode.

In addition to common clothing materials, textile sensors can also be made by combining some synthetic flexible polymer materials with carbon-based materials [9]. Research on the application of conductive nanofiber composites in ECG monitoring systems has been lacking. In 2021, Huang et al. developed a stretchable, flexible nanofiber-membrane carbon electrode that can be used for ECG monitoring. The carbon electrode was composed of rGO, CB, and PU. The nanofibers were made by mixing PVDF and poly (3, 4-ethylene dioxythiophene) polystyrene sulfonate (PEDOT/PSS) using electrospinning technology. Electrospinning offered a fast and easy way to prepare nanofibers [10]. During electrostatic spinning, the polymer solution was charged at high pressure, causing the solvent to evaporate, and subsequently deposited on the collector as a fiber. The fibers prepared by electrospinning were nanoscale and had a high specific surface area. The nanofiber carbon electrode has high conductivity, water resistance, and durability, which is suitable for long-term physiological-signal monitoring [11].

2.2. CNT/CB-Based Textile Electrodes/Sensors for ECG Monitoring

Flexible sensors based on CNTs generally show a large sensing range but low sensitivity or high sensitivity but a small sensing range [12][13]. To solve this problem, Dong et al. developed a synergistic CB–CNT-combination stretchable conductive network and superhydrophobic perfluorooctyltriethoxysilane-modified TiO_2 nanoparticles (PFOTES-TiO_2 NPs) in 2022. They designed and fabricated an ultra-stretchable self-cleaning non-woven textile bioelectrode with excellent health monitoring and antifouling performance. The preparation process involved dipping the Styrene-ethylene-butylene-styrene (SEBS) fabric spun by electrodeposition into a mixture of CNTs and CB, followed by brushing the mixture of PFOTES, ethanol, and TiO_2 to form a superhydrophobic layer on the surface of the textile.

3. Carbon-Based Textile Sensors for Body Motion Monitoring

In general, human motion signal monitoring can be divided into large movements such as joint and limb movements and small body deformations such as in the pulse and breathing [14]. Most carbon-based textile sensors that monitor human motion are flexible strain sensors. There has been a lot of research that focuses on the development of carbon-based strain sensors for physiological-signal monitoring. For more details, readers may consult the following recent reviews [15][16]. Strain sensors work by converting signals of mechanical deformation into electrical signals. According to different principles, they can be divided into resistance, capacitance, and piezoelectric sensors. The resistive sensor structure mainly consists of a conductive sensitive film and flexible substrate. The change in the conductive pathway caused by the strain leads to the corresponding change in resistance. The capacitive sensor structure is mainly composed of a flexible medium in the middle and conductive electrodes on either side of the medium. One main sensing mechanism of capacitive sensors is the deformation resulting in a change in the volume of the space between the electrodes and thus, a change in the capacitance of the sensor [15]. Piezoelectric sensors are mainly made of piezoelectric materials. The measurement principle is that stress causes lattice deformation of piezoelectric materials, which leads to the change of the crystal polar state. Therefore, as the piezoelectric material strains, it generates an internal electric field that converts pressure into electricity. Piezoelectric materials can be divided into four categories: ceramics, single crystals, polymers, and composites (composed of piezoelectric ceramics or single crystals in a polymer matrix) [17].

3.1. Graphene-Based Textile Sensors for Body Motion Monitoring

Graphene is used to make strain sensors to monitor human movement. The combination of graphene with flexible textiles is also worth investigating. Yarns with sensing properties not only have a low cost and simple manufacturing process; they can use weaving technology to form a better sensing structure, and the measurement position is more flexible [18][19]. In 2017, Li et al. reported the preparation of a novel yarn strain sensor based on commonly used PU yarns that were easy to integrate into textile structures and apply textile technology to wearable applications. The yarn was wrapped with a graphene/polyvinyl alcohol composite material as a conductive sheath and polyurethane yarn as an elastic core. A simple, scalable, and low-cost layered assembly method was adopted. By combining the advantages of graphene, such as superior electrical and mechanical properties and thermodynamic stability [20], with the advantages of polyurethane yarn, such as high tensile strength and elasticity, and good textile processing properties [21], a strain sensor with high sensitivity and good tensile strength was produced. It can be seen that the sensor can almost maintain the good tensile strength and elasticity of its polyurethane core, including high tensile strength and linearity, as well as low hysteresis. When the strain is about 40%, the stress–strain curve of the sensor almost coincides with the stress–strain curve of the polyurethane core. When the strain exceeds 40%, the stress–strain curve of the sensor deviates only slightly from that of the core. This shows that coating the polyurethane core with graphene nanosheets does not significantly change the elasticity of the polyurethane yarn while providing high sensitivity [22].

Huang et al. applied a simple spin-coating technique to apply GNPs to textiles to make strain sensors and used polyaniline (PANI) particles in GNPs to improve sensor performance. In 2017, they prepared a textile strain sensor for human gesture recognition based on the highly conductive PANI polymer, GNPs, and silicon rubber (SR). the structure of the textile strain sensor mainly includes the PDMS encapsulation layer, the conductive composite material (GNPs/PANI/SR), and the elastic PU fabric electrode. The resistance-sensitive material of the sensor is the GNPs/PANI/SR film. The addition of PANI particles improves the electrical conductivity of the thin film. The small-sized PANI particles are attached to the surface and edge of the GNPs. When the sensor is stretched, PANI particles fill the gaps generated by the stretched GNPs, not only maintaining the structure but also forming a collaborative conductive network with GNPs. This results in an increase in electrical conductivity. Five separate strain sensors were implanted into the spandex gloves to detect the bending and stretching of the fingers.

To further improve the sensitivity of sensors, Huang et al. reported a polymer nanosphere decorated with graphene-composed porous fibers for wearable and highly sensitive strain sensors in 2019. Inspired by rolling friction, the group reduced the interconnections between graphene sheets and polymers by reducing the contact area. They designed porous graphene fibers (PGFs) with polymer nanospheres mixed with PVDF and PU inserted between the sheets This made the PGFs highly sensitive while being used for long durability. Pulse is a very important but relatively small physiological signal. This sensor can be used to monitor pulse waves. The pulse signal was very clear with a beating frequency of 68 min^{-1}. The insertion plot (black curve) showed a close-up of the peak of a single pulse and produced the typical characteristics of the pulse waveform, i.e., shock wave (P wave), tidal wave (T wave), and relaxation wave (D wave). When the vein was pressed, the signal strength (red curve) was lower than the normal signal and the pulse frequency dropped to about 60 min^{-1}. This indicated the high sensitivity of PGFs to distinguish small changes [23].

3.2. CNT-Based Textile Sensors for Body Motion Monitoring

CNTs are widely used in the manufacture of strain sensors because of their good electrical conductivity and stability. In 2018, Liu et al. proposed a new method to fabricate strain-sensing fabrics by embedding single-CNT strain-sensing yarns into textile structures. The yarns were coated with polyvinyl alcohol (PVA) to obtain higher mechanical properties. The CNT-based yarns coated with PVA had better durability and mechanical properties. They performed with stable, repeatable, and linear sensing properties in the cyclic loading process. The CNT-based textile sensor showed a fast and accurate response to finger motion detection, demonstrating the potential of wearable electronics [24].

To improve the air permeability and comfort of fabric sensors, different methods have been used to make flexible sensors. In 2019, Doshi et al. used an innovative and scalable method of electrophoretic deposition to deposit CNTs onto knitted fabrics to develop a flexible, lightweight, and comfortable tensile sensor for human motion monitoring. Knitted fabrics consisted of 44% nylon, 43% polyester, and 13% spandex. Due to the chemical bonding of the CNTs to the fiber surface, the robustness of the coating ensured a repeatable response over multiple test cycles.

The high sensitivity and wide sensing range of strain sensors are usually contradictory. The strain sensors' formation by the electrostatic spinning of sensing yarn can effectively improve their performance. In 2020, Qi et al. created a CNT-embedded PU nanofiber sensing yarn (CNTs@PU-NF). It was a stretchable, multimodal, wearable textile strain sensor made by adding CNTs to elastic PU nanofibers. First, stretchable, conductive, carbon-rich nanotube fibers were fabricated as core fibers and electrodes. CNTs were then added to elastic PU nanofibers and wrapped on stretchable core electrodes by one-step electrospinning to form stretchable piezoresistive nanofiber sensing yarns. Finally, these nanofiber sensing yarns served as warp and weft yarns that were perfectly integrated into the wearable textile substrate using weaving technology. The sensor responded immediately to minute pressures with a fast response time of 30 ms. The sensors with nanofiber sensing layers of different thicknesses showed a monotone sharp increase in resistance with strain along different GF values and had a wide strain sensing range. The hysteresis of the sensors was negligible when the strain was applied back and forth between 0% and 40%, and only slight hysteresis was observed at tensile strains greater than 220%. The mean GF value of the sensors was 114 in the 100% strain range and 720 in the 100–220% strain range. Even at a tiny strain of 0–1%, they had a significant GF value of 54.9. The CNTs@PU-NF sensors showed high cyclic stability and reproducibility and can be used to monitor human daily movements [25].

3.3. CB-Based Textile Sensors for Body Motion Monitoring

CB is also used in the production of strain sensors because of its good electrical conductivity and low cost. Due to the inevitable inelastic deformation of the textile, response fluctuations and hysteresis still exist [26]. To address this limitation, Luo et al. created a flexible piezoresistive sensor (FPS) in 2018. CB particles and PVDF were added into knitted fabrics as electrical and mechanical interconnections between the fibers. The knitted structure was interwoven yarn of twisted, conductive nylon fibers. The knitted fabric was decorated with CB grains. PVDF was introduced to improve the adhesion of CB to fibers. At the same time, it can form an interconnection between the fibers, thus filling the air gap and reducing hysteresis deformation. A CB/PVDF film uniformly covered the fiber surface and indeed formed interconnections between fibers in many areas. The sensor can capture subtle physiological-signal changes, such as deep inhalations and deep exhalations. The sensor can also be used to monitor superficial temporal artery pulse pressure and pulse wave velocity [27].

Although the method of coating CB directly on the flexible textile is simple and low-cost, it has low tensile strength and sensitivity. Therefore, it requires further research and exploration of diversified textile structures with higher performance. In 2021, Park et al. designed a strain sensor with high tensile strength and sensitivity using low-cost carbon-based ink. The carbon-based ink was mainly composed of CB, supplemented by gelatin and PU mixed, with characteristics of being waterproof, stable, and washable. Two-dimensional triaxial-braided weaving was used to obtain this textile, which was soaked in a carbon-based ink solution and dried so that enough CB was evenly attached to the surface of the textile. The initial resistance was measured at 20 kΩ. The carbon ink-coated textile was pre-strained 130% at a rate of 10 mm/min. In practical application, the cracks separate during tension and the resistance increases rapidly, thus enhancing the sensitivity of the sensor with GF up to 80. After the release of external force, the textile also returns to its original state. The existence of cracks also makes the sensor with air permeability suitable for long-term physiological-signal monitoring. The doping ratio of PU affects the adhesion between the carbon black and the fabric. Fabrics coated with a PU content of 10 wt% (PU-10) show excellent performance in the test. The initial resistance remains unchanged before and after washing. It also shows high stability and long-term durability after 5000 cycles of tensile testing. After testing, the sensor can not only monitor the physiological information of low-scale movements such as breathing or pulse, but can also obtain the physiological information of large-scale movements such as joint movements [28].

3.4. Graphene/CNT-Based Textile Sensors for Body Motion Monitoring

In addition to performance, the durability and washability of the sensors for daily use needs to be considered. CNTs have poor adhesion to textiles by coating or inkjet printing. In 2020, Tang et al. reported a machine-washable electronic textile sensor with high strain-resistance and high thermal conductivity. CNTs were fixed to non-woven fabric by the nano-soldering method, and then rGO was combined with the CNTs/NWF. The textile exhibited excellent properties. When the tensile strain was 1.0%, the strain coefficient was 32.65. It could detect subtle strains as low as 0.01%. The textiles showed excellent washability. After washing in a washing machine with a stirring rate of 700 rpm, the electrical resistance and thermal conductivity did not change much. At the same time, both rGO and CNTs were retained on the NWFs, and the carbon weight did not change significantly. The textile was used to measure human movement. For example, it was worn on fingers to record finger movements. The higher the curvature of the fingers, the greater the change in conductance is. When the finger returns to its original position, the conductance correspondingly returns to its original value. It can also be mounted on the hand to monitor pulse rate and can also record different human pulse waveforms (diastolic, tidal, and shock peaks) [29].

Nano-soldering methods can improve the adhesion of conductive materials on textile surfaces and can be also used in the fabrication of strain sensors. In 2021, Yao et al. reported highly sensitive gas-permeable strain sensors that are washable and wearable. The team created a sensor based on a mixture of graphene and CNTs on a non-woven fabric (NWF) substrate by using the ultrasonic nano-soldering method. The electronic textile had high electrical conductivity, was highly sensitive to tensile strain and pressure, and can be used as a wearable sensor for medical monitoring. The wearable rGO/CNTs/NWF sensors can monitor human movement by being worn on the index finger. When bending to different states, the sensor shows significant changes in resistance, and when the finger returns to the same position, the resistance values are nearly identical. In addition, the wearable rGO/CNTs/NWF sensors were used to detect pulse, muscle, and heart rate. They can record distinct human pulse waveforms (diastolic, tidal, and percussion peaks) at a pulse frequency of 12 per 10 s (72 beats/min), within the normal range for healthy adults. Significant pulse signals can be detected, and signal peaks are significantly dense. They can still maintain high electrical conductivity after machine washing. These results indicate that wearable rGO/CNTs/NWF sensors have excellent repeatability, washability, and durability and are sensitive to both weak physical stimuli and large movements; therefore, they have great potential applications in smart clothing, personal healthcare, human movement, and robotics [30].

To deal with the complicated manufacturing method of textile sensors, Zhou et al. reported a textile strain sensor based on screen printing technology to transfer graphene-nanosheets (GNSs)/NWCNT mixed ink to cloth tape. It has good linearity and stability with low manufacturing costs. The sensing range and sensitivity of the textile sensor can be adjusted according to the different design methods, which can be used in human motion monitoring such as gesture recognition [31].

4. Carbon-Based Humidity Textile Sensors for Respiration Monitoring

Breathing can provide some information for the diagnosis of lung disease. The relative humidity of exhaled air is less affected by environmental factors such as temperature and movement. Therefore, the development of flexible humidity sensors for real-time continuous respiration monitoring is one of the most popular research topics. In 2022, Xing et al. reported a humidity sensor made of textile coated with MXene and MWCNT for real-time respiration monitoring. The researchers used alternate-drip coating to form MXene and MWCNT layers over a superfine fiber fabric (MC-Fabric). After MWCNT layers were inserted, the distance between MXene molecule layers became wider, which was conducive to the adsorption of more water molecules [32]. The response rate of the sensor was 265% at 90% relative humidity. The humidity response of the MC-Fabric sensor varied by less than 3% under both folded deformation and torsional deformation. The MC-Fabric could be integrated into a facemask to accurately identify various human breathing patterns. One subject was tested for four representative breathing patterns including normal breathing, rapid breathing, slow deep breathing, and apnea. The breathing curve was visible, and different breathing patterns were clearly distinguished. The experimental results showed that the MC-Fabric sensor has good responsiveness and excellent repeatability, which provides a feasible method for real-time respiratory monitoring based on electronic textiles [33].

5. Carbon-Based Textile Sensors for Body Temperature Monitoring

Body temperature is often the simplest and most straightforward indicator of a person's health. Many diseases are accompanied by changes in body temperature. Researchers in the field of flexible sensors have been working to achieve long-term continuous temperature measurements. The application of carbon-based materials in textile sensors for body temperature monitoring has also been studied. In 2021, Arman et al. developed a CNT-based temperature textile sensor. The conductive CNT-based ink was prepared by inkjet printing and deposited on Taffeta fabric. The surface was coated with a translucent PU film as a protective layer. The sensor showed a negative temperature coefficient characteristic with which the resistivity of the sensor decreases with the increase in temperature. In addition, it can be used to measure human body temperature in real-time conditions [34].

6. Carbon-Based Tactile Textile Sensors

As an important way of perception, touch can make people respond differently to external mechanical stimuli. The research on tactile sensors is important, and carbon-based materials are also used in the production of tactile sensors. Bae et al. used inkjet printing technology to prepare a highly sensitive wearable CNT-based tactile sensor in 2022. It is difficult to form high-resolution patterns on textiles with printing techniques such as screen printing [35] and dip coating [36], which are commonly used in the production of textile sensors. To solve this problem, they used inkjet printing technology. Uniform CNT-based patterns were formed on cotton textiles, achieving a variety of high spatial resolutions of up to 0.5 mm patterns.

7. Carbon-Based HR and SpO_2 Sensors

HR and SpO_2 are important parameters of human health, and it is very important to monitor them for a long time, which can be applied to monitor diseases and exercise conditions. HR and SpO_2 are usually measured by photoplethysmography (PPG). PPG is favored by researchers for non-invasive physiological-signal monitoring through the optical monitoring of blood flow [37][38]. In 2019, Polat et al. made a GOD (graphene sensitized with semiconducting quantum dots) photodetector. Because of its transparent nature, graphene can be used to make wearable photoelectric sensors. It uses flexible wearable devices and integrates with pre-designed electronic components. It can detect vital signs that need to be tracked continuously for a long time, and it can work under ambient light [39].

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