Carbon Nanotube-Polyaniline Reinforced Flexible and Stretchable Strain Sensor with Enhanced Sensitivity for Smart Wearable and Skin-Mounted Applications

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ABSTRACT: Flexible and stretchable strain sensors have emerged as promising components for integration into smart wearable devices and skin-mounted applications. These sensors enable accurate detection of physiological signals, thereby finding unique applications in diverse fields such as human health monitoring, soft robotics, human-machine interface, prosthetics, virtual reality, and professional sports. Two commonly utilized types of strain sensors are capacitive and resistive strain gauges, owing to their low production cost, simplified circuitry, and ease of construction. While resistive strain gauges exhibit high sensitivity, they are prone to nonlinearity and hysteresis. On the other hand, capacitive strain gauges demonstrate linear behavior with minimal hysteresis but offer lower sensitivity. In this study, we capitalize on the exceptional properties of carbon nanotubes, including high mechanical strength, electrical conductivity, and thermal stability, along with using polyaniline as an exemplary conductive polymer. These materials are employed as a reinforcing phase within the polymer matrix, while the dielectric layer is comprised of Ecoflex® 00-30. An interdigitated pattern is specifically designed for this strain gauge to enhance sensitivity. Through this research, we aim to develop a flexible and stretchable strain sensor with enhanced sensitivity and improved performance characteristics.

KEYWORDS: gauge factor, strain sensor, tensile strength, wearable flexible.

INTRODUCTION

With the gradual advancement of digital health technologies in recent years, the field of electronic smart wearables has experienced rapid growth, utilizing a combination of nanomaterials and nanocomposites. Given the increasing demand for flexible electronic devices, particularly strain sensors and skin-mountable sensors in wearable industries, numerous projects have been undertaken in this field for various potential applications, including personal health monitoring, human motion detection, human-machine interface, and soft robotics. One of the key driving factors behind the progress in this field is the essential need for digital healthcare to monitor patients’ conditions away from hospitals remotely. Strain sensors are devices that convert mechanical deformations into electrical signals. Flexible and stretchable strain sensors are mainly classified into resistive [1-4], capacitive [5-7], and piezoelectric [8-9] types. However, capacitive and resistive strain sensors have simpler readout circuits, greater flexibility, and higher stretchability. Resistive strain sensors consist of a thin conductive layer adhered to an elastic substrate. When this composite structure is stretched, microstructural changes in the thin layer lead to a change in electrical resistance. This change is a function of the applied strain. Upon releasing the stress and returning the strain sensor to its initial state, the electrical resistance of the thin layer will also return to its initial state [10]. A capacitive strain sensor exhibits a change in capacitance in response to applied strain, demonstrating an inverse relationship. A capacitive strain sensor consists of a dielectric sheet sandwiched between two electrodes whose thickness can vary with respect to the applied strain. Tensile and compressive strain cause the electrodes to move closer to each other, resulting in an increase in capacitance. A capacitive strain sensor with parallel electrodes typically includes a backing layer for strain measurement and pressure detection. However, its adaptation to irregular surfaces such as human skin is challenging. On the other hand, a capacitive strain sensor with interdigitated electrodes exhibits better sensitivity, linear behavior, and lower hysteresis [11].

Due to their metallic structure and the brittle nature of thin semiconductor layers, traditional strain sensors were relatively rigid and had limited stretchability. As a result, they lacked the capability to be mounted on the skin or fabric. These sensors were constructed from rigid materials such as silicon, which required high-temperature processes, making their application in smart
weareables and soft robotics challenging. Therefore, achieving flexibility and increasing the strain range is of great importance for these sensors.

To address this issue, nanomaterials, and nanocomposites have been employed to form the optimal structure of wearable strain sensors with good mechanical and electrical properties. These sensors are based on nanomaterials and composed of a combination of nanomaterials within a flexible and elastic substrate, such as fiber, fabric, or polymer. Various processes for their production are utilized, including spray coating, layer-by-layer deposition, dip coating, and various printing and lithography techniques. Lately, we have successfully created nanocomposites by combining PLGA with natural substances, making them highly promising contenders for producing composites based on polymer/carbon nanotubes [12]. A nanocomposite-based strain sensor typically consists of two crucial components: a conductive network that provides a pathway for signal conduction or transmission and a polymer elastomer part for flexibility, stretchability, and protection of the conductive network. Achieving a strain sensor with high sensitivity and stretchability requires a logical selection of materials and structural design. PLGA/Fibrin composite fibers demonstrate significant potential, stimulating the growth and proliferation of adipose-derived cells while also influencing the tensile strength of the composite [13].

Among the significant advancements made in the fabrication of wearable strain sensors, there are still numerous challenges in this field. The first challenge is the selection of suitable materials. All conductive nanomaterials, such as carbon nanomaterials, metallic nanowires, and conductive polymers, have their limitations. However, finding ways to mitigate the weaknesses of these materials and produce a good conductive network with high adhesion capability is an essential initial issue. The second challenge is achieving high stretchability, high sensitivity, and linear behavior simultaneously in a strain sensor. High stretchability requires a faultless morphology under significant strain, high sensitivity necessitates substantial structural changes, and linear behavior requires homogeneous morphology during stretching. The third challenge involves scalability and the cost-effectiveness of the sensor for commercialization. To achieve mass production, designing a manufacturing process based on full printing can significantly contribute to cost reduction and commercialization efforts.

Recently, strain sensors based on graphene have been reported. However, due to the brittleness of graphene, they exhibit low stretchability. Additionally, dual elastomer-nanowire and nanoparticle structures have been reported, but due to their weak adhesion to the flexible substrate, they are not suitable for constructing these strain sensors. Ultimately, strain sensors based on liquid metals and ionic liquids have been reported, demonstrating good stretchability (over 550%) and low hysteresis but low sensitivity (less than 5) [14]. For a capacitor with two parallel plates of initial length \( l_0 \), width \( w_0 \), and dielectric layer thickness \( d_0 \), this equation \( C = \frac{\varepsilon_0 \varepsilon_r (\varepsilon + 1) \omega d_0}{d_0} \) is correspond (Where \( \varepsilon_0 \) and \( \varepsilon_r \) represent the permittivity of free space and the relative permittivity, respectively) [15]. For a capacitive strain gauge with interdigitated electrodes which consists of overlapping length \( l_0 \), width \( w_0 \), initial thickness \( t_0 \), and distance between the two electrodes \( d_0 \), we have \( C = \varepsilon_0 \varepsilon_r \frac{(1-\omega)+1}{(1+1)d_0} \) (n-1) (which \( n \) is the number of electrodes). For the design of a high-performance strain gauge, criteria such as sensitivity, linearity, stretchability, response and recovery time, hysteresis, drift, dynamic durability, creep behavior, production cost, ease, and simplicity in production and packaging play a fundamental role [16, 17].

In this research, carbon nanotubes have been used as fillers in the elastomeric matrix network due to their high electrical conductivity and desirable mechanical properties. Carbon nanotubes exhibit excellent electrical conductivity, and due to weaker van der Waals bonds in carbon nanotubes, the process of dispersion is relatively simple, and the tendency for agglomeration is reduced, allowing for a uniform distribution within the elastomeric matrix network.

**MATERIALS**

In order to achieve outstanding electrical and mechanical properties, we integrated carbon nanotubes into a strong polymer matrix consisting of polyaniline, renowned for its exceptional conductivity. Additionally, we utilized Eco-flex 0030 to enhance the composite's stretchability. The carbon nanotubes utilized in this study were multi-walled, boasting a purity level exceeding 95%, and possessing an outer diameter ranging from 20 to 30 nm. These nanotubes were acquired from US Research Nanomaterials, Inc. in Houston, USA. Other essential materials, such as Aniline, Ammonium Peroxoydisulfate, HNO3, H2SO4, and HCl, were procured from Merck (Merck Pte. Ltd., Darmstadt, Germany).
EXPERIMENT AND RESULTS

Considering that carbon nanotubes do not have a natural tendency to form bonds due to their non-reactive nature, they need to be functionalized first, and their outer surface needs to be made accessible. Therefore, the process of carboxylating carbon nanotubes was carried out to create COOH functional groups on the carbon nanotubes [18, 19]. In the next step, polymerization is performed, and a polymer coating is applied to the carbon nanotube filaments. This not only improves the mechanical properties of the carbon nanofibers but also prevents their agglomeration [20]. The main electrode material is a combination of carbon nanotubes and polymers. Due to the brittle and dry nature of this combination, it requires flexibility for use in a flexible sensor. Therefore, to achieve this goal, through multiple experiments and trial and error, it was found that by combining this material with Ecoflex 00-30 in a ratio of 1:2, a flexible electrode with suitable electrical conductivity can be obtained. Additionally, this method reduces the amount of the main material used in the sensor, leading to a lower overall sensor cost.

The design was initially created using Corel software to create an interdigitated pattern mold. In this design, the distance between the electrodes is 500µm, the length of the electrodes is 20mm, the thickness of the electrodes is 1mm, and the depth of the electrodes is 200µm. The material for mold fabrication used in this method is a 5mm thick polycarbonate sheet. After loading the interdigitated pattern into the laser engraver, the surface of the polycarbonate sheet was engraved.

The sensor materials, including a combination of carbon nanotubes, polymer, and Ecoflex 00-30, are filled into the engraved sections of the template using the doctor blade method. Then, the unengraved areas are covered with adhesive labels to ensure that the surface of the polycarbonate sheet is completely free from sensor materials. Subsequently, the polycarbonate sheet is placed inside a rectangular hollow cube mold with a thickness of 6mm, which is 1mm thicker than the main template. This mold is designed for pouring Ecoflex onto the main template in a way that, upon pouring the Ecoflex onto it, the layer thickness reaches 1mm. After the Ecoflex has dried, which takes approximately one day, the peel-off method is used to separate the Ecoflex layer. To ensure good electrical contact, Agar conductive copper tape was used to connect the contacts. Additionally, to enhance the adhesion between the copper tape and the contact surface, carbon adhesive with a resistance of approximately 32Ω was applied. Therefore, a small piece of thin copper foil, about 3 centimeters in size, was separated and adhered to the electrode contact area using the liquid carbon adhesive. After approximately 15 minutes, the adhesive dried.
Figure 1. a. Fabricated capacitive strain sensor. b. Graph of variations in length, strain, capacitance, and changes in capacitance relative to the initial capacitance over time in a strain gauge with 20 pairs of serrations at a strain of 2%. c. Graph of changes in length, strain, capacitance, and variations in capacitance relative to the initial capacitance over time in a strain gauge with 20 teeth at a 20% strain.
Figure 2a. The relative changes in capacitance, relative to the applied strain of 2%, for the 20-pair toothed capacitance strain gauge are illustrated in the graph. 

Figure 2b. The relative changes in capacitance, relative to the applied strain of 20%, for the 20-pair toothed capacitance strain gauge are illustrated in the graph.

Figure 3a. The relative changes in capacitance, relative to the applied strain of 2%, for the 20-pair toothed capacitance strain gauge in the strain and release (hysteresis) states are depicted in the graph.

Figure 3b. The relative changes in capacitance, relative to the applied strain of 20%, for the 20-pair toothed capacitance strain gauge in the strain and release (hysteresis) states are depicted in the graph.
As we know, the capacitance of an interdigitated strain sensor decreases during the tension phase and increases during the release phase. The decrease in capacitance during the tensile state, perpendicular to the direction of the electrodes, is due to the increased distance between the electrodes. We initially subjected the capacitance strain gauge with 20 pairs of teeth to a 2% strain. This strain gauge exhibits a good sensitivity to this small strain and responds accordingly, as shown in Fig 1 b. We evaluated a capacitance strain gauge with 20 pairs of teeth at a 20% strain. This strain gauge demonstrates the ability to respond to a 20% strain, as shown in Fig 1 c, with its capacitance decreasing as the strain increases and increasing as the strain decreases.

The sensitivity of the capacitance strain gauge factor is equivalent to the slope of the relative capacitance change versus the applied strain. Fig 2a represents the relative capacitance changes of the strain gauge in relation to an applied strain of 2%, where the slopes of the curves correspond to the gauge factor of the strain gauges, which is 12 for the strain gauge with 20 pairs of teeth. In theoretical calculations, assuming equal Poisson's ratios for the dielectric layer and electrodes, the gauge factor of the strain gauge is not dependent on the geometric shape and the number of teeth. Furthermore, the nearly linear behavior of these strain gauges can be observed in the graphs for an applied strain of 2%, indicating a good response of the sensor within this range. The strain gauge factor with 20 pairs of teeth at a strain of 20% is shown in Fig 2b. As evident, this strain gauge exhibits completely linear behavior only up to a strain of 5%, and its gauge factor has also reduced by 1/4 due to the decrease in capacitance variations at higher strains.
The relative capacitance variations with respect to an applied strain of 2% for a 20-tooth strain gauge in both the loading (blue line) and unloading (red line) phases are illustrated in Fig 3a. As evident, the blue line indicates the applied strain (loading path), and the red line represents the unloading phase (return path). The strain gauge exhibits good performance with very minimal hysteresis. This is in contrast to resistive strain gauges, where hysteresis is a significant challenge. The hysteresis of the strain gauge with 20 pairs of teeth at 20% strain is shown in Fig 3b. It can be observed that there is almost no hysteresis in this sensor, and the behavior during the tension phase in the strain gauges corresponds to their behavior during the release phase.

In order to observe the drift characteristics of the sensor, a static strain of 10% was applied to the strain gauge for a duration of 100 seconds and maintained in this state. It can be observed in Fig 4 that under the static strain of 10%, the capacitance of the sensor does not undergo significant changes throughout the stretching period.

In order to better understand the performance of this strain gauge, we attached it to the surface of the wrist skin. As indicated in the capacitance versus time in Fig 5, bending the wrist leads to a decrease in the capacitance, and returning to the initial state restores it to its original value. Additionally, for applications that require strain gauges with smaller dimensions, such as finger movements, it is possible to utilize strain gauge samples with fewer electrodes. In future research, we want to incorporate the carbon nanotube-based capacitive strain gauge into the microfluidic device's design. When the microfluidic device is subjected to mechanical strain, caused by pressure changes or fluid flow, the strain will also impact the carbon nanotubes, allowing for real-time monitoring of mechanical strain on the microfluidic device.

CONCLUSION

Based on the main objectives of this research, which were to achieve a flexible strain gauge sensor with linear behavior, negligible hysteresis, and a high gauge factor, we were able to develop a cost-effective strain gauge using suitable nanomaterials and simple methods. This strain gauge exhibits a gauge factor of 12. While numerous studies have been conducted on capacitive strain gauges, most of them demonstrate linear behavior and negligible hysteresis, but their gauge factor typically remains around one, which is within the theoretical range. However, due to the high gauge factor of our strain gauge at low strains, it can be used in applications that require detecting small strains, such as respiratory issues, small body movements, and overall human health monitoring. In addition to its ability to be attached to the skin, it can also be mounted on fabric surfaces. The strain gauge, being encapsulated with Ecoflex 00-30, is capable of withstanding large deformations and various strains. Furthermore, this strain gauge has the capability to respond to higher strains up to approximately 20% applied strain, although its sensitivity decreases with increasing strain. Nevertheless, the level of hysteresis remains negligible and acceptable.

REFERENCES


11. Wearable and Transparent Capacitive Strain Sensor with High Sensitivity Based on Patterned Ag Nanowire Networks-2017


15. A Highly Elastic, Capacitive Strain Gauge Based on Percolating Nanotube Networks-2012

16. Recent Developments in Bio-monitoring via Advanced Polymer Nanocomposite-based Wearable Strain Sensors- 2018


