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Low-cost ultra-stretchable strain sensors for monitoring human motion and bio-signals

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ABSTRACT

The emerging need for interactive wearable devices has been a driving force for the development of flexible electronics. Due to their ability to conform to the complex nature of the human physique, stretchable strain sensors have been extensively used to measure bio-signals and monitor human motion. Here, a new fabrication method of a piezo-resistive strain sensor is introduced, and the ability of the sensor to measure the human heartbeat and track a wide range of human motion is demonstrated. The sensor is fabricated by infusing graphene nano-flakes into a rubber-like adhesive pad. The fabricated sensor is highly stretchable and can withstand strain up to 350%. Even after 10,000 cycles of stretching and relaxing a robust and stable electrical response is maintained. Furthermore, the gauge factor of the sensor ranges from 2 to upto 160; which in turn allows the sensor to monitor a great variety of human motions. Hence, three wearable devices are developed using the fabricated graphene-based strain sensor to measure human knee movement, finger movement and heartbeat through the radial artery. The sensor is also used in a robotic haptic application to control a robotic finger. These experiments demonstrate the applicability of the sensor for real-time monitoring, specifically in wearable human interactive devices.

1. Introduction

Recent advances in wearable devices have shown substantial promise for applications in health monitoring and biomedical fields. On the other hand, miniaturizing electronic technology has enabled researchers to introduce ad hoc wearable devices for certain applications. However, realizing the full potential of wearable devices for commercial purposes still requires advancements in sensitivity, stretchability and flexibility [1–5] of the sensing elements to easily conform to broad varieties of human physique and respond to large and small joint movements, such as bending and rotating. Consequently, flexible electronics are accepted as an excellent candidate to be used in wearable devices causing less discomfort while measuring human bio-signals and physiological activities [6–9].

Stretchable strain sensors are a specific type of flexible electronics that allow precise measurement of deformation in wearable devices. There have been many endeavours to make highly stretchable and flexible strain sensors that are more compliant with human body specifications and have reasonable size, sensitivity, performance, production cost, and robustness in various environments [10–14]. In that regard, stretchable and flexible strain sensors should be sensitive enough to respond accordingly to different and complex body motions that may range from infinitesimal movements and bio-signals (e.g., heartbeat) to large stroke muscle movements (e.g., walking and running) [1,15,16].

Strain sensors are generally categorized into three groups: piezo-voltage, piezo-capacitive, and piezo-resistive. Piezo-voltage strain sensors are effective at detecting small strains due to their large gauge factors (GFs). However, they are not inadequate for human motion monitoring because they are not sufficiently stretchable and cannot withstand large strains (<30%). [5,17,18] Piezo-capacitive strain sensors have a more linear response and a lower hysteresis but have a lower gauge factor (GF < 1). [1,19,20] Among the three major strain sensor groups, piezo-resistive strain sensors have received the most attention due to both their simplicity and their tunable mechanical and electrical properties. However, the low gauge factor (GF < 2) [21–23], low range of tolerable strain ($\varepsilon < 5\%$) [15,24], and high brittleness (Young's modulus > 1×10^{10} Pa) [25] of the previously reported piezo-resistive

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strain sensors hindering their full functionality in wearable devices. To better facilitate the interaction of piezo-resistive strain sensor with the human body, their sensing ability and mechanical compliance has been improved through recent use of various materials and fabrication techniques. These include implementation of nanoscale conductive films on flexible substrates, development of metallic nanocomposite structures [26–28], and most recently, the use of carbon nanotubes [23,29–31], nanowires [21,32–34], and graphene nano-powder on flexible substrates [35–39]. Despite these recent advancements, stretchable strain sensors require further development to meet the necessary criteria for conforming to complex human motions. As previously mentioned, this criteria includes reasonable sensitivity and stretchability, robustness in various environments and durability as well as easy fabrication process and low production cost [11,40–42].

The aim of this study is to address the current lack of adequate flexible electronics technology for versatile wearable devices by fabricating a new stretchable strain sensor and applying it to interactive human-machine interface applications. This was accomplished by developing an easy fabrication method for a low cost, highly stretchable piezo-resistive strain sensor (capable of detecting a wide range of strain) and utilizing it for both human motion monitoring and haptic implementation. The sensor was fabricated by infusing graphene nano-flakes (GNFs) into an adhesive pad and exhibits superior mechanical properties that can withstand more than 350% strain. With our particular fabrication method we made a graphene-based strain sensor with an initial resistance as low as 8 kΩ and a gauge factor as high as 161. The sensor has a linear response below 40% strain and exhibits very low hysteresis up to 100% strain. The fabricated sensor is also omnidirectional and is sensitive to different types of deformation including bending, twisting, compressing and stretching. To utilize the sensor for human motion monitoring, we embedded the sensor in an Ecoflex® substrate (a widely used skin compatible polymer). The sensor is capable of detecting strains as small as a human pulse and as large as human knee movement; since the sensor also has a fast recovery time, this enabled us to measure various human physical activities (e.g. walking and running).

2. Results and discussion

A fabrication method for development of a new stretchable and flexible strain sensor has been achieved by infusing GNFs into nano-pores of a commercially available rubber-like adhesive pad (Re-POP™ Clear Adhesive Pad) (Fig. 1). Initially, the pad was placed into an acetone bath; this resulted in the pad to undergo volumetric expansion and increased to over twice its initial size. This increased the pore size of the pad and facilitated the infusion of GNFs into the substrate. Next, a solution of GNFs, methanol, and water (GMW) was prepared; the solution was mixed in a sonication bath to ensure uniform dispersion of GNFs. To infuse GNFs into the swollen adhesive pad (AP) the pad was placed into the GMW solution. The solution was left in a sealed container at room temperature for 6 h without any agitation. The pad infused with GNFs (GNF-Pad) was removed from the solution and dried overnight at room temperature. The GNF-Pad was washed to remove residual graphene particles and expose the strain sensor's smooth, black, and bright surface.

To characterize the quality of the sensor's specifications, each step in the aforementioned fabrication process was analysed (Fig. 1). The rubber-like adhesive pad swells when submerged in different organic solvents, including acetone and chloroform. Acetone was selected for the solvent in the fabrication process since it is less hazardous than chloroform. Various soaking times in acetone were examined (Fig. 2a) to characterize the pad's swelling behaviour; we found that the pad swells to its maximum size (2.3 times its initial volume) after 5 h. Furthermore, soaking in acetone (upto 48 h) did not affect the mechanical properties of the AP, both in terms of the Young's modulus and ultimate tensile strain after it dried (Fig. 2b and c). Interestingly, the adhesive property of the pad was unaffected by soaking in acetone, and the pad recovered its adhesive property when removed from the bath and dried. The pad's adhesive property restricts the infused GNFs movement; the flakes stick to the polymer substrate and cannot freely slide within the medium. Thus, they are moved apart increasing the resistance when the polymer substrate stretches. Consequently, this strain sensor is very durable and does not require any additional elements to create adhesion between GNFs and the substrate.

During the second step of the fabrication process the swollen AP was submerged in the GMW solution. Due to differences in osmotic pressure, the acetone exited the pad and GNF particles entered through the expanded pores. During this phase the pad shrunk to its original size. To ensure smooth deposition and linear shrinking of the pad, the GMW solution composition was optimized. We examined the swollen AP's deformation during shrinking when placed in GMW solution with different ratios of water to methanol (100%:0%, 80%:20%, 75%:25%, and 50%:50% water to methanol ratios, respectively) (Fig. S1). A ratio of 75%:25% water to methanol displayed linear shrinking of AP without any bending; this ratio was selected to create the GMW solution as it guaranteed uniform deposition of the GNFs on the surface of the AP. Although optimizing the GMW is important to ensure uniformity of the sensor, the final quality largely depends on the AP soaking time in the GMW solution. As such, the effect of soaking time was measured and the maximum depth of infusion was found to be approximately 13 μm, and achieved after 6 h soaking (Fig. 2d). The initial resistance R_0 (defined as resistance without any applied strain) of the GNF-Pad for different soaking times was also assessed (Fig. 2e). The R_0 is directly related to the soaking time; resistance of the GNF-Pad soaked for 30 min was 150 kΩ/cm but dropped to 8 kΩ/cm when soaked for 6 h. Since longer soaking times did not significantly change the resistance of the pad (Fig. 2e), a 6-h soaking time was used to fabricate sensors for further study.

To achieve a uniform and robust GNF layer on the pad, the GNF-Pad was removed from the GMW solution and dried at room temperature overnight. The water content of the GMW solution in the soaked pad, lowers the evaporation rate and provides more time for the GNFs to orient and uniformly deposit on the substrate while drying. This creates a bright and smooth layer of GNFs on the substrate. The sensor was washed with deionized water after drying to remove residual GNFs on the substrate.

A stress-strain curve was conducted to assess the effect of the fabrication process on the mechanical properties of the AP and the final sensor (Fig. 2f). Both samples displayed very similar stress-strain curves suggesting the process had a minimal effect on the mechanical properties. Scanning electron microscope (SEM) images taken from cross-sections of the GNF-Pad revealed a uniform infusion of the GNF (Fig. 2g–i). The GNF also were in close proximity with one another and had continuous distribution (Fig. 2i). Collectively these characteristics give the sensor a low initial resistance.

The response of the fabricated strain sensor was examined for various levels of strain (Fig. 3a). The sensor was capable of withstanding 350% strains without any observable cracking or other signs of failure. The sensor's response up to 40% strain was linear and the resistance increased to 3 times its initial significant, this may not be a major concern in the application of the proposed sensor in wearablvalue of 8 kΩ ($\Delta R/R_0 = (R - R_0)/R_0 = 3$ at 40% strain $\Delta R/R_0 = (R - R_0)/R_0$; R_0 is the initial resistance of sensor with no strain, Fig. 2e). Further increase in strain caused an exponential increase in the relative resistance ($\Delta R/R_0$): at 350% strain