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Stretchable and compressible strain sensors for gait monitoring constructed using carbon nanotube/graphene composite

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Abstract

Stretchable and compressible strain sensors play an essential role in various fields with uses ranging from automotive components to medical devices. This study reports on the fabrication and characteristics of stretchable strain and pressure sensors constructed using a carbon nanotube and graphene composite. The sensors were used for gait analysis, an important step in the diagnosis and management of movement disorders. The stretchable and compressible strain sensors were used to measure peak knee sagittal angles and forces under the feet when walking. Gait analysis is usually performed within a laboratory. However, in this research we propose a shift to gait assessments conducted via long-term daily monitoring using wearable devices.

Introduction

The growing demands in highly stretchable and deformable materials for use in human–machine interfaces [1] and human motion detectors [2] have motivated the development of highly sensitive stretchable strain and pressure sensors with high strain tolerance. Several stretchable strain sensors have been prepared using nanomaterials coupled with stretchable polymers [3–9]. Stretchable polymers such as poly(3, 4-ethylenedioxythiophene) doped with poly(styrene sulfonic acid) (PEDOT:PSS) [3], thermoplastic polyurethane (TPU) [4], natural rubber [3], low density polyethylene (LDPE) [6], polydimethylsiloxane (PDMS) [7, 8], and polyurethane (PU) [9] are generally selected as flexible matrix composites. Among these, PDMS is a good candidate due to its outstanding elasticity, good thermal stability, transparency, and stable chemical properties [10]. CNTs are easy to entangle due to the large aspect ratio resulting in low fracture toughness [11] and electrical property of the composites [12]. One of the simple and effective ways to reduce the agglomeration is to incorporate CNTs with graphene to produce a nanocomposite material or a hybrid [13]. Hence, to determine the advantages of novel preparation methods, these nanostructures were used as fillers in this research. In table 1, the proposed sensor is compared with other CNTs composite strain sensors.

In this work, we propose a novel method for fabricating stretchable strain and pressure sensors using the diffusion of polymer through a nanostructure network. This method is simple and inexpensive and offers a plethora of opportunities to produce various types of sensor compositions. In this research, PDMS was used as the matrix polymer and a CNT-graphene mixer was the nanoscale network. The sensitivity of the sensor can be controlled by the CNT and graphene composition ratio. Both stretchable strain and pressure sensor capabilities of the sensors were tested for gait analysis application.
Increasing human longevity brings increased degeneration of the musculoskeletal and nervous systems, increased incidence of Parkinson’s disease and stroke, and more people suffering from walking difficulties. Gait disorders lead to reduced mobility, concomitant development of weakness, loss of independence, and decreased quality of life [21]. However, gait analysis has seen only limited usage because of the high cost involved in setting up a gait laboratory. Human gait analysis is useful both for identifying the underlying cause of an abnormal gait and for gait physical therapy.

Figure 1. Fabrication process of the stretchable and compressible strain sensors.
Experiments

Materials
PDMS and a curing agent were purchased from Dow Corning Co. CNTs (Nanogeneration Co, Thailand) have a purity of about 99.99%, length of 3–12 um, outer diameter of 12 nm and the wall thickness of 4 nm. Nanographene platelets (NGPs) with a documented average size (x and y) of <10 μm, thickness of <10 nm, were purchased from Angstron Materials, Dayton, OH, U.S.A. Hexanes (RCI Labscan) was used as received without any further purification.

Preparation of the CNTs/graphene composites sensors
The fabrication procedure for CNT/graphene composite-based strain sensors is schematically shown in figure 1. CNTs and graphene at varying mass ratios (0.4 g:0 g, 0.3:0.1 g, 0.2 g:0.2 g, 0.1 g:0.3 g and 0 g:0.4 g) were

Table 1. Comparison of working range and gauge factor of previous reported strain sensors based on graphene/CNTs/PDMS composite.

| Materials                   | Highest GF | Strain range | References |
|------------------------------|------------|--------------|------------|
| Ag&CNTs / PDMS              | 20         | 2%           | [14]       |
| CNTs/Nonwoven               | 5.34       | 1.8%         | [15]       |
| CNT&GO / PDMS               | 1.6        | 80%          | [16]       |
| CB/PDMS                     | 5.5        | 10%          | [17]       |
| MWCNTs/PDMS                 | 1.2        | 45%          | [18]       |
| CNTs & graphene / PDMS      | 0.36       | Not provided | [19]       |
| CNTs & Graphene / PDMS      | 11.4       | 9%           | [20]       |
| CNTs & Graphene / PDMS (this work) | 11       | 80%          | —          |

Figure 2. Photograph of the cross-section of the stretchable strain sensors.
separately dispersed in 20 mL ethanol and sonicated for 15 min and subsequently stirred gently for 2 h at room temperature. Then the two solutions were mixed together for another 2 h of stirring. The final mixture was poured into a 30°30°5 cm³ poly (methyl methacrylate) (PMMA) mold. After the evaporation of the solvent, CNT-graphene agglomerations were produced. After that, a 10:1 mixture of PDMS and curing agent was stirred thoroughly and cast on the CNT-graphene agglomerations. PDMS diffused through the layer of CNT/graphene

Table 2. The average GF and the average maximum strains of four composite samples.

| Samples   | Average GF | Average maximum strain (%) |
|-----------|------------|-----------------------------|
| C100      | 3.3        | 60                          |
| C25:G50   | 4.5        | 41                          |
| C50:G50   | 10.9       | 71                          |
| C75:C50   | 4.8        | 50                          |

Figure 3. Photograph of the complete of the stretchable strain sensors (a), relative change of resistance versus strain of the C100:G0, C25:G75, C50:G50 and C25:G75 (b)–(e), respectively. 
Figure 4. SEM image of CNTs (a), graphene (b), PDMS (c) and CNTs-graphene PDMS composite sensor.

Figure 5. The repeatability test under strain of 33% for 1000 cycles.
agglomerations to produce CNT-graphene composites. After drying for 2 d at room temperature, the samples were peeled off the mold and cut to the desired size.

**Characterization**

The electrical resistance of the bending and stretching tests was measured with a multimeter (Keithley 2450). The stretchable strain sensors were used to produce kneepads to measure the bending of the knee while walking, squatting, and running.
Result and discussion

As shown the sensor cross-section in figure 2, the sensor consisted of 2 layers: a CNT-graphene composite with approximate thickness of 0.4 mm and PDMS with a thickness of 1.5 mm. Furthermore, the inset picture shows that the composite is on just one side of the sensor and its thickness is about one-fifth of the sensor’s thickness. Due to the large amount of virgin PDMS in the sensors, the flexibility and stretchability of the sensor is similar to that of pure PDMS.

The strain sensor’s gauge factor (GF) represents the sensitivity of the sensor and is defined as

\[ GF = \frac{\Delta R}{R_0} \frac{R_0}{\Delta L/L_0} \]

where \( \Delta L/L_0 \) is the strain on the sensor and \( \Delta R/R_0 \) is the relative change in the resistance of the sensor.

Figure 3 shows optical photographs of a 1 cm width strain sensor. Copper wires were bonded using silver glue on the two ends of the CNT/graphene composites, the distance between electrical contact was 8 cm. Then, PDMS was cast on top of the Ag electrical contact to produce a sandwich structure, as shown in figure 3(a). The relative changes of the sensors with varying CNT:graphene ratios, CNTs 100%, CNTs75% - graphene 25%, CNTs50% - graphene 50%, CNTs25% - graphene 75% (C100, C75:G25, G50:G50, C25:G75), and the sensor resistances are shown in figures 3(b)–(e). The graphene 100% without CNTs was an insulator; therefore it was not included in this experiment. In order to ensure the reliability of the data, at least three of each sample with the composited size of 1” x 8” x 0.04 cm was tested. The behavior of the samples with the same composition showed similar performance. It can be seen that the \( \Delta R/R_0 \) of all the samples increased with the increase of applied strain. The average initial resistances of the CNT-graphene/PDMS composite strain sensor were 4.7 kΩ, 4.9 Ω, 91.2 Ω and 108.2 kΩ for the C100, C75:G25, C50:G50, and C25:G75, respectively. The sensors with large amount of CNTs (C100 and C75:G25) exhibit low initial resistance and sensitivity due to a great deal of electrical contact points. The C100 strain sensors can be stretched up to 60% with average sensitivity of GF = 3.3 (shown in figure 3(b)). The C25:G75 show average GF of 4.5 with the strain limit of 41% (figure 3(c)). As shown in figure 3(d), the C50:G50 exhibit the highest GF of 10.9 and the average maximum strain was 71%. The average GF of the C25:G75 is found to be 4.8 with the average maximum strain of 50%, figure 3(e). These results are due to during the process of stretching/releasing, the conductive network is destroyed and reconstructed leading to changes in resistance [22, 23]. The experiment shows that C50:G50 exhibited the best performance for strain sensing because at that ratio, the conductive network can be destroyed easily, so higher sensitivities are obtained. Since it had the highest GF, this sample was chosen for further study. Table 2 summarizes the average GF and the average maximum strains of four composite samples.

The morphologies of the CNTs, graphene, PDMS and CNTs-graphene PDMS composite are shown in figures 4(a)–(c). The diameter and length of the CNTs used in this research were approximately 100 nm and 2–3 μm, shown in figure 4(a). Figure 4(b) showing several micron-size graphene sheet. Figure 4(c) is SEM image.
of the composite, it was much rough compared with the pure PDMS. The graphene sheets were observed all over the surface. Furthermore, CNTs were found spread throughout the matrix.

Durability is an integral characteristic of a wearable strain sensor due to the high levels of strain of human movements. Degradation of electromechanical performance due to plastic deformation or breakage of active materials is not desirable. One end of the sensors was attached to a rotating disk which rotated at a period of 16 s. Figure 5 shows the mechanical durability of the sensors when stretched sinusoidally at strain ranging between 0%—33% for 1000 cycles. The relative change of resistance slightly decreased after the first 100 cycles and then stabilized. It dropped by less than 7% by the completion of 1000 cycles of the durability test, demonstrating good stability.

To demonstrate the application of the CNT-graphene composite sensors in gait monitoring, we attached the sensor to a knee, as shown in figure 6(a). When walking forward, the knee joint bends and straightens. The corresponding resistances were measured. The \( \Delta R/R_0 \) increases as the knee bends (the sensor is stretching) and decreases as the knee joint moves back to its straightened state (figure 6(b)). As shown in figure 6(c), the sensor also successfully measured running movements.

Figure 9. Photograph of the electrical connection with attached to CNTs-graphene composite (a), diagram of the pressure sensor (b), pressure sensor.
To use the stain sensor as a wearable device, we designed a knee band (1 cm x 8.5 cm x 0.19 cm) to be worn with the strain sensor at Patella position, as shown in figure 7. The sensor was stretched at least 100 times before use to detect the bending. The system is controlled with an Arduino nano microcontroller with a built in microSD card slot and is powered by a 3.7 V, 2000 mAh rechargeable lithium ion battery. The microcontroller recorded data to the SD card as a volunteer neurologist shuffled like a Parkinson’s patient. Figure 8 shows the voltage measured across the sensor. We found that, both amplitude and frequency signals from normal walk and shuffling gait are completely different. Normal walk revealed high amplitude and low frequency whereas shuffling gait showed low amplitude and high frequency. It is clearly seen that the device successfully classified patients with shuffling gait.

The sensor prepared in this report is not limited to detecting strain, but it can also be used to sense pressure. For that purpose, the composites were cut to the size of 1 x 1.5 cm, and one-third of the area was used as the electrical contact area. As shown in figure 9(a), the copper wires were attached by silver glue on the composites. Next, they were articulated together to create a force sensor, the schematic diagram of which is shown in figure 9(b). Finally, the sensors were put in a 3.5 x 2.5 x 0.7 cm mold and cast by pouring PDMS. The complete pressure sensor is shown in figure 9(c).

To evaluate the performance of the pressure sensor, it was placed under the heel of a 50 kg woman and stepped on. Figure 10 shows the resistance of the sensor during the press and release. The resistance in the range of 3.5–7 kΩ demonstrates the sensor’s high stability.
The pressure sensor was used to produce a data shoe to monitor gait in the same way as the strain sensor. Arduino nano, microSD card slot, and 3.7 V lithium ion battery were used again within the same objective. The sensor was placed under the foot and the voltage across the sensor were recorded (figure 11 and inset). It was found that normal walk and shuffling gait revealed the roughly same amplitude but different frequency. Moreover, we can see that the signals when running are larger than those when walking.

Conclusion

We presented a novel method to fabricate stretchable and compressible strain sensors using the diffusion of polymer through nanostructure materials. The fabrication strategy was simple and inexpensive and produced high sensitivity. The strain sensor had a high GF (10.9) and high durability (more than 1000 cycles). Moreover, the pressure sensor was stable under high pressure (30 kg cm⁻²). Furthermore, a data knee band and data shoes were built using the stretchable and compressible sensors to classify the disorder gait.

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