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FULL PAPER

Wearable and Highly Sensitive Graphene Strain Sensors for Human Motion Monitoring

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Sensing strain of soft materials in small scale has attracted increasing attention. In this work, graphene woven fabrics (GWFs) are explored for highly sensitive sensing. A flexible and wearable strain sensor is assembled by adhering the GWFs on polymer and medical tape composite film. The sensor exhibits the following features: ultra-light, relatively good sensitivity, high reversibility, superior physical robustness, easy fabrication, ease to follow human skin deformation, and so on. Some weak human motions are chosen to test the notable resistance change, including hand clenching, phonation, expression change, blink, breath, and pulse. Because of the distinctive features of high sensitivity and reversible extensibility, the GWFs based piezoresistive sensors have wide potential applications in fields of the displays, robotics, fatigue detection, body monitoring, and so forth.

1. Introduction

Graphene is a two-dimensional and structurally hexagonal honeycomb material. Under tensile load, the structure of the hexagonal honeycomb would partially be destructed near the edge of the film.[1] It would result an alteration of the electronic band structure and conductance characteristics, subsequently, causing a significant change of the resistance of the graphene.[2–5] The combination of amounts of extraordinary properties, such as ultra-translucency, superior mechanical flexibility and stability, high restorability, and carrier mobility, enables an application of graphene in high-sensitivity strain sensors,[6,7] which can be used in future displays,[4,8] robotics,[9,10] electronic skin,[11] in vitro diagnostics,[12–14] and human physiological motion detections.[10,15–17] Monitoring human physiological signals has been considered as an effective approach to evaluate human health even at the current age of highly-developed medical technology. In order to directly obtain various strain levels of the human physical motion, recently, researchers have focused on developing a new technology of putting high-sensitive nanostructured materials in human bulk geometries. The stain sensors at high gauge factors strain sensors, based on ZnO nanowires autologous vertical arrays or polymers, have exhibited promising pressure sensing properties of fast response of about 140 ms. The sensors may be used in nano-sensor systems for precision measurements.[15,18] In comparison with the rigid metal or metal oxide nanowires, carbon nanotubes (CNTs) exhibits distinct advantages and features, such as excellent strain gauging performances, good conductivity, high-transparency, superior physical robustness, and easy fabrication. Thus, CNTs have potential applications in bio-interactive and intelligent electronics.[10,16,17] In addition, it was reported that a device with micrometer-scale patterned arrays, which were formed by casting with polydimethylsiloxane (PDMS) or metal-coated (like Pt) rubber on the pretreated Si moulds, with special tiny structures can respond to weak motion signals, such as pulse or heartbeat more easily and precisely.[14,19–21] Despite the excellent pressure sensing performance, it is still a grant challenge to detect human motion signals by the sensors because of the complex structures, difficultly fabricated, high-cost, and sophisticated equipment needed.

In this work, a simple-structured and low-cost graphene woven fabrics (GWFs)[22] strain sensor, which can readily distinguish various strain levels of human motion signals, was prepared. As the stress was applied on the strain sensor, high-density cracks generated in the network, leading to the decrease of current pathways decrease and the increase of resistance. Because of this special crisscross configuration, the GWFs possessed an extremely high gauge factor, which is calculated to

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be ≈10^3 under 2–6% strains, 10^6 under higher strains (>7%),[23] and ≈35 under small strain of 0.2%. It indicated that the sensor could be stretched by a tiny deformation of 0.2% with obvious resistance change, enough to be recorded. The signals of any weak motions, including breathing, phonation, expression changes, blink, and pulse, can be detected. On the other hand, it could endure a large deformation of 30% with the completely reversible electrical properties. In order to mould around human skin well to ensure the response of real signals, a good adhesive medical tape with PDMS glue, which is a flexible, biocompatible, shape controllable material,[24] upside as the substrate was used. As a new kind of electronic skin, it was made up with graphene woven fabrics, PDMS, and medical tape. The device was wearable and biomedical compatible enough to be placed inside clothing or mould around human skin without any irritating feelings.

2. Results and Discussion

Figure 1 shows the schematic illustration of the key processes in fabrication and operation of a strain sensor based on GWFs-PDMS-medical tape film. GWFs were obtained by atmospheric pressure chemical vapor deposition (CVD) method[22] through graphene growth on the surface of crisscross copper meshes, similar with the graphene growth on the copper foil substrate. Subsequently, the copper meshes were etched away in $FeCl_3$/HCl solution. GWFs were transferred to a pretreated film composited with medical tape and PDMS. The GWFs-PDMS-tape was then dried and connected to silver wires with silver paste on both ends to obtain the final GWFs based strain sensor. The thickness of the medical tape/PDMS composite was about 0.5 mm. The sheet resistance of the GWFs was about 400–500 Ω sq^{-1}. Another PDMS glue was used to cover the electrodes for the protection from exfoliation. The multiple devices were then finally assembled.

Figure 2a presents a photo image of an integrated detector assembled by GWFs-PDMS-tape of about 6 cm × 6 cm. As seen in the image, the mechanical property of GWFs was strong enough to prepare a large-scale sheet fabric. The wearable devices can be easily tailored to pieces with appropriate size for sensing applications. A magnified optical microscope image is shown in Figure 2b. The structure of the original cylindrical graphene obviously collapsed onto the PDMS plat and formed a double layered mesh film (Figure S1, Supporting). Figure 2c illustrates the high adhesion of human body at different parts, demonstrating the wearable and flexible features of the monitoring devices. Because of the strong van der Waals force between GWFs and PDMS and the piezoresistive effect of graphene, it was believed that graphene strain was synchronizedly generated with the PDMS-tape substrate.[25,26] The novel graphene networks in this work exhibited excellent performance under different tensile strains. Its relative resistance change could be 10 times at 2% strain or 10^4 times at 8% strain,[22] and also could be 0.07 time at 2‰ strain. The change is large enough for ordinary instruments to detect the motion signals, as shown in Figure 2d.

As shown in Figure 2c, the sensor moulds around human skin well because of the high quality of the medical tape. To detect the high sensitivity of the GWFs films for human motion detection, the device was used to monitor slight movements of human, such as hand motions from stretch to clench, phonation, change in facial expressions, blink, breathing, and even pulse. Large movements, such as elbow motions and knee motions, have not been described here because graphene resistance changes are more obvious under large deformation. Our previous work[22] provided the measuring results about large strain of GWFs for different finger motions. The resistance changed significantly in the hand motion from stretch to clench, as shown in **Figure 3**a, if the multiple devices were equipped on the back of a hand. The inset shows about two different level of strength when hand clenched. As expected, when

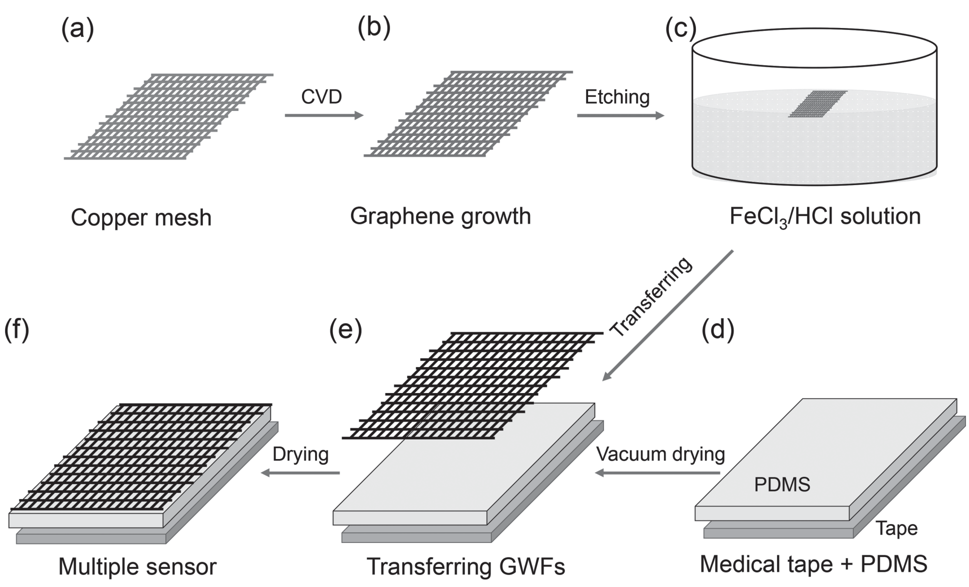

Figure 1. Schematic illustrations of fabrication procedure of a human motion sensor based on GWFs-PDMS-medical tape film.

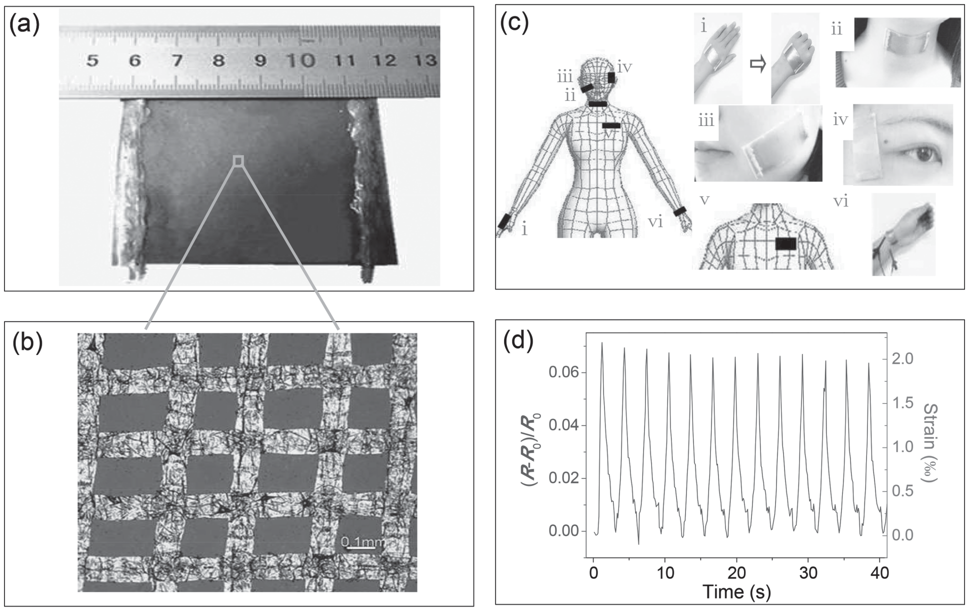

Figure 2. a) Optical photograph of a GWFs-PDMS-tape composite film; b) A magnification of the optical microscope image; c) Photo images of GWFs-PDMS-tape at various positions of; d) Relative change of resistance between 0 and 0.2% strain.

the greater strength was applied, the deformation of muscle was larger, the collected signals were more evident. Figure 3a shows an enlarged view of the yellow frame marked in the illustration. Peaks represent hand motions with fast response. The details of an in situ tensile test with the real time monitor of resistance change when hand moving from stretch to clench are shown in Figure S2 and Supplementary Movie 1. The wearable and highly sensitive sensor even could be attached at soundtrack directly, as shown in inset image of Figure 3b, to distinguish out different signals of muscle motions when a tester spoke different words or phrases, such as “Hi”, “Hello”, “CNMM”, and “NanChang University (NCU)”. The yellow frame 2 is an amplified view of yellow frame 1. It can be seen that waveform of single curve was apparently different and the repeatability of each curve was very good. By combining the magnitude of the resistance change with tensile performance, in Figure 2d, the deformation strain caused by phonation vibration can be speculated about 0.1~0.2%.

The GWFs-PDMS-tape multilayered detector is suitable for monitoring different facial expressions as well. The results of changes of facial expression and blinks are shown in **Figure 4**a,b, respectively. Each single peak corresponded to a facial expression change from poker face to smile or a blink. The recorded signals of GWFs resistance change was attributed to GWFs deformation, which was caused by internal facial muscle stretch corresponded to expression changes or blinks. If GWFs could closely fit with the tester’s whole face, the sensor might be applied as a lie detector based on the muscle deformation amplitude in the future. The blink test also could be potentially used in evaluation of the degree of fatigue or human health easily by measuring the frequency and amplitude of blinks. Furthermore, the signal detected from GWFs-PDMS-tape based sensor was stronger than that for ZnO (the current change was 5 nA at 30% strain or the voltage change was ≈50 mV at 0.1% strain)[15,18] and CNTs (the resistance change was 2% at weak motion test when the sensor adhered to the throat)[10,27] based sensors.

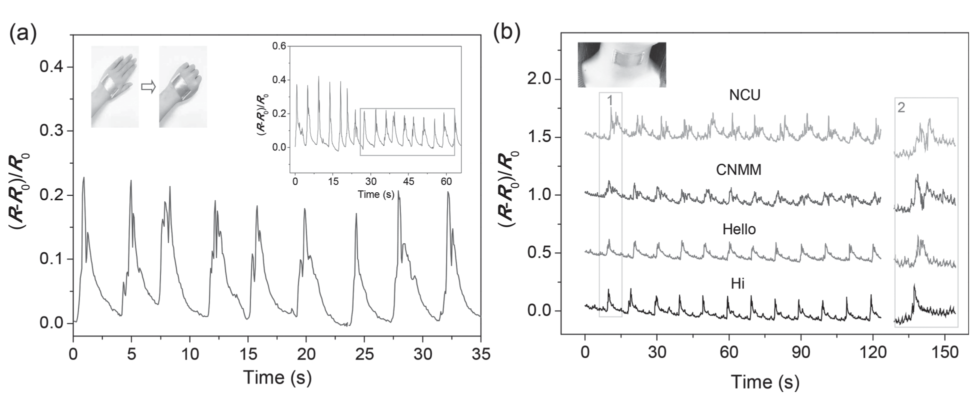

Figure 3. a) Relative change of resistance in hand motion from stretch to clench; b) Real-time relative change of resistance in the muscle motions in speech.

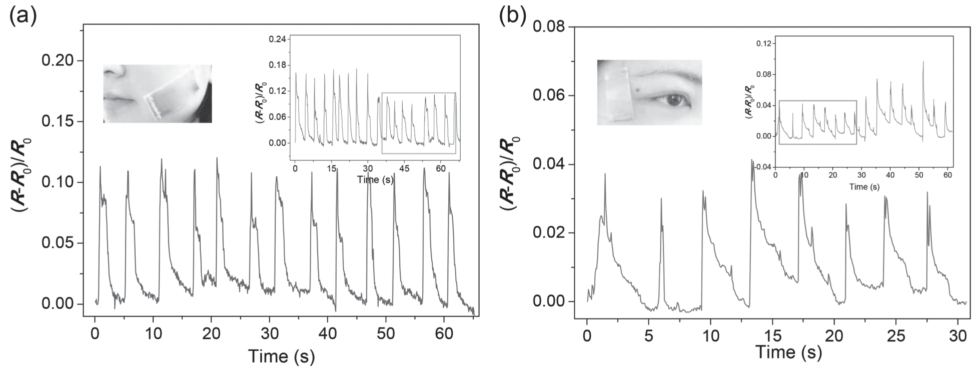

Figure 4. Relative change of resistance in muscle motions of expression changes and blinks, in (a) and (b), respectively.

As vital signs, respiratory and pulse are important to be detected in a hospital. GWFs-PDMS-tape sensor can be used to take those vital signs under both still state and movement state. The signals of the breathing rate and pulse rate are shown in **Figure 5**a,b, respectively, at two body states of still (black) and exercise (red). Each cycle represents a breath or a pulse. The peaks and valleys are assigned to the chest stretch and shrink, respectively, in breathing. The results completely complied with the realistic physiological behaviors. The signal of exercise state was of higher frequency and larger amplitude than that of still state. As shown in Figure 5a, the breath numbers in still state and exercise state are 32 and 55, respectively, in 100 s. The pulse numbers in still state and exercise state are 38 and 46, respectively, in 30 s. All data is within the normal range of a healthy adult.

3. Conclusion

By using a crisscross copper mesh as the substrate, highly sensitive GWFs were fabricated by the CVD method. A flexible and wearable strain sensor was assembled by adhering the GWFs on PDMS and medical tape composite. The sensor exhibited the following features: ultra-light, relatively good sensitivity, high reversibility, superior physical robustness, easy fabrication, ease to follow human skin deformation, without irritation, and so on. As a consequence of the piezoresistive effect of graphene woven fabrics, the sensors were used as electronic skin covering human body to detect body motions. The signals of GWFs resistance change depend on deformation strain which is formed by the motions. The stronger the motion is, the larger the strain is, and the easier the motion signals can be recorded. Some weak human motions were chosen to test the notable resistance change, including hand clenching, phonation, expression change, blink, breath, and pulse. Because of the distinctive features of high sensitivity and reversible extensibility, the GWFs-PDMS-tape based piezoresistive sensors have wide potential applications in fields of the displays, robotics, fatigue detection, body monitoring, in vitro diagnostics, and advanced therapies.

4. Experimental Section

Synthesis of GWFs: Copper meshes (100 mesh, wires of 100 micrometers in diameter) were cleaned, tailored, and pretreated as reported previously.[22] When the temperature reached 1000 °C, H_2 was turned down to 20 cm^3 min^{-1}. CH_4 (30 cm^3 min^{-1}) was introduced into the reactor at ambient pressure. And Ar was cut off for 1 min, and then turned up to 200 cm^3 min^{-1}. After 20 min growth, the mesh was rapidly cooled down to room temperature. Graphene was grown around the copper mesh. GWFs could be obtained when copper mesh was etched away by an aqueous solution of $FeCl_3$ (0.5 mol L^{-1}) and HCl (0.5 mol L^{-1}).

Fabrication of GWFs-PDMS-Medical Tape Strain Sensor: The fabrication of PDMS/medical tape film is described as follows: The PDMS mixture of base and cross-linker (the weight ratio of base to cross linker was 10:1) was stirred at least for 20 min. The medical tape was attached evenly on a clean disposable plastic petri dish, and then PDMS mixture was spin-coated onto the medical tape gently, degassed in vacuum for 10 min to remove bubbles at room temperature, solidified at 80 °C for 1 h, and

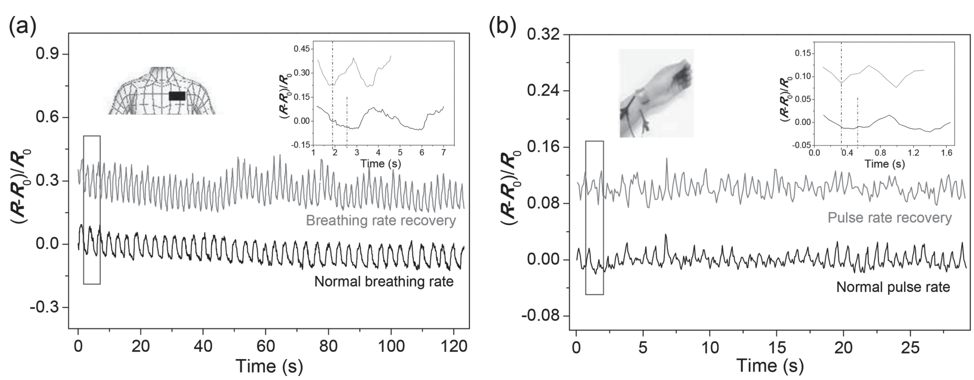

Figure 5. Relative change of resistance of the respiratory and pulse, in (a) and (b), respectively, at still state and exercise state.

peeled off from the petri dish to get the composite PDMS-medical tape film (≈0.5 mm thick). After transferring and drying the GWFs on PDMS-medical tape film, silver wires were connected using silver paste to form the GWFs-PDMS-medical tape strain sensor. PDMS glue was used for covering the electrodes to protect them from exfoliating, and then the multiple devices were assembled.

Characterizations: The samples were characterized using an optical microscope (Axio scope A1) and scanning electron microscopy (Leo1530). The tensile test was carried out on a stretching machine (Instron 5943). The electrical properties was recorded by a digital source meter (Keithley 2602 and Keithley 4200-SCS), and the Source-Drain voltage with 1 V DC bias.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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