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Ultra-sensitive graphene strain sensor for sound signal acquisition and recognition

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ABSTRACT

A wearable and high-precision sensor for sound signal acquisition and recognition was fabricated from thin films of specially designed graphene woven fabrics (GWFs). Upon being stretched, a high density of random cracks appears in the network, which decreases the current pathways, thereby increasing the resistance. Therefore, the film could act as a strain sensor on the human throat in order to measure one's speech through muscle movement, regardless of whether or not a sound is produced. The ultra-high sensitivity allows for the realization of rapid and low-frequency speech sampling by extracting the signature characteristics of sound waves. In this study, representative signals of 26 English letters, typical Chinese characters and tones, and even phrases and sentences were tested, revealing obvious and characteristic changes in resistance. Furthermore, resistance changes of the graphene sensor responded perfectly with pre-recorded sounds. By combining artificial intelligence with digital signal processing, we expect that, in the future, this graphene sensor will be able to successfully negotiate complex acoustic systems and large quantities of audio data.

1 Introduction

Sound is a vibration that propagates as a typically audible mechanical wave of pressure and displacement through media such as air, water, or solids, and its perception and processing are integral parts of human intelligence. Traditional sound collection and recognition involve the process of taking samples of an analog sound and storing the results in storage media or as binary data. Humans can use auditory

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sensors to receive external sounds through vibration of the spreading air. However, when one cannot physically produce vocalizations due to some disease, how could others know what one wants to say? Therefore, it is necessary to develop a new device (and attendant method) for collecting and recognizing communication through signals other than ordinary vocalizations. Accordingly, sensing strain and vibration with soft materials at small scales has garnered increased attention recently.

Vocalization signals are characterized according to their observable output [1]. Phonological or prosodic features of the voice include loudness, jitter, fundamental frequency, zero-crossing rate, and energy frequency ratios [2]. Normally, audio recorders or computer recording software (such as voiceprint) can collect and recognize the sounds of human speech almost perfectly through the greatly reduced spectral information. However, such processes severely restrict the listener's ability to obtain complete information according to the distribution of spectral energy [3]. When someone experiences difficulty in producing or hearing a sound, a commercially available artificial electronic throat [4] or electronic ear [5, 6] can be used to either produce or collect and recognize the sounds of speech.

Recently, it has been shown that another type of strain sensor attached to the throat could record muscular movements (Fig. 1) in order to collect and recognize speech sounds. This is permitted by the fact that the throat muscle exhibits different degrees of stretching or shrinking strains when speaking different words [7, 8]. Sensitivity, flexibility, stretchability, and softness are therefore important attributes for a throat sensor of this sort [9–12]. In strain sensors [13–15], the gauge factor (GF) is defined as the change in resistance with respect to the mechanical deformation, and this quantity therefore expresses the sensitivity of the material. More specifically, the relationship between the change in electrical resistance (dR/R) and the variation of mechanical strain (dL/L) is given by $GF = (dR/R)/(dL/L)$, which indicates that the strain

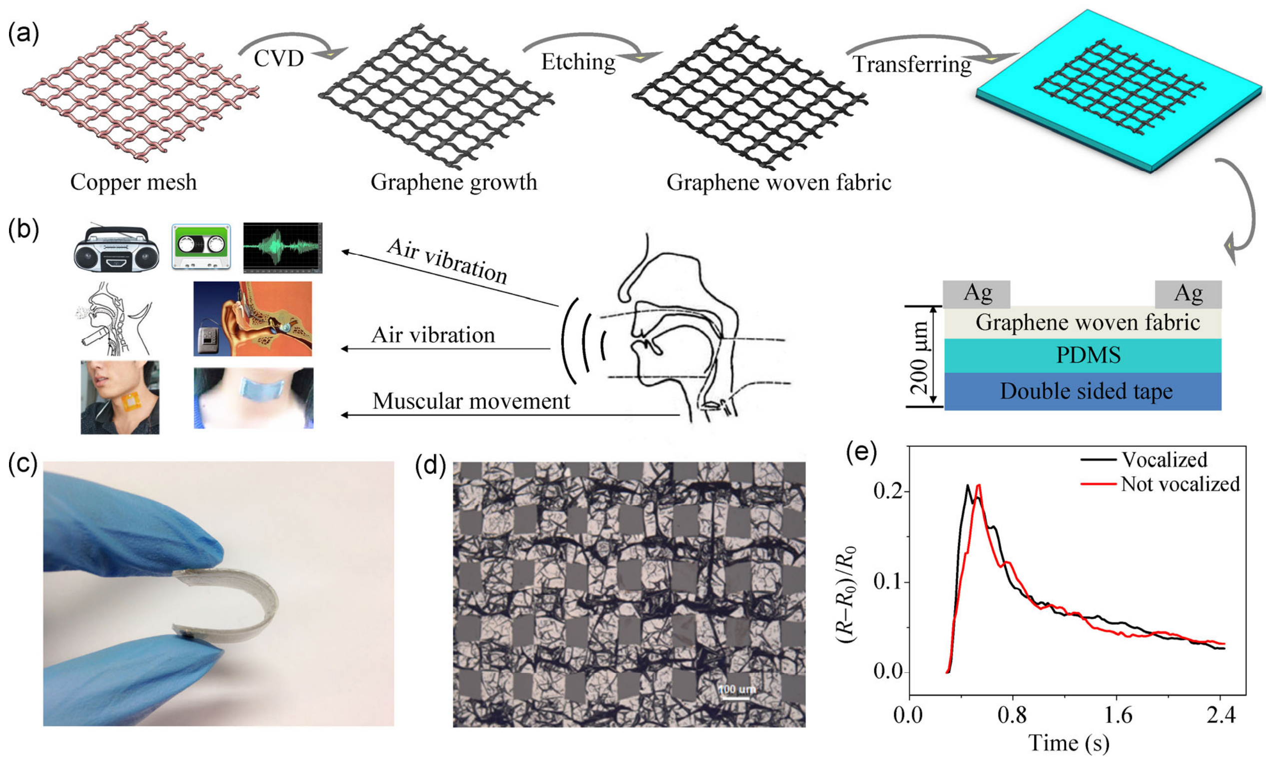

Figure 1 Graphene woven fabric on polydimethylsiloxane (GWF-on-PDMS) strain sensor. (a) Key steps of preparing the graphene strain sensor. (b) Three ways of collecting and recognizing human voices. (c) Photograph of a bent strain sensor. (d) Scanning electron microscopy (SEM) image of GWFs. (e) Signals of vocalization (black) and non-vocalization (red) nearly overlap when a tester performs the same speech action.

sensor with a larger GF value is more sensitive [16]. To list a few examples, it has been reported that the GF of carbon nanotube (CNT)/polymer composite is 0.06–0.82 [17]; that of traditional metal is 1–5 [18]; that of a semiconductor is ~100 [19]; that of a CNT-based sensor is ~1,000 [20]; and that of a single nanowire-based sensor is ~1,250 [21]. The GF of graphene woven fabrics (GWFs) from our research group can be as high as 10^3 with 2%–6% strains, 10^6 with higher strains (> 7%), and ~35 with a minimal strain of 0.2%. These GF values are the best yet recorded, and are measurable with a normal digital multimeter. Additionally, due to their high sensitivity, excellent flexibility, and lack of irritation to human bodies, GWFs have been used as strain gauges, the key component in many monitoring sensors (e.g., finger bending, hand clenching, expression change, and breath and pulse monitoring) [8, 22, 23].

In this study, we demonstrate one interesting example of the potential applications of GWFs in high-sensitivity detectors—namely, the wearable acoustic sensor. GWFs (crisscross graphene) prepared by chemical vapor deposition (CVD) on copper meshes were transferred onto the thin, biocompatible, elastic polymer polydimethylsiloxane (PDMS) with double-sided tape [24, 25]. When stress was applied on the GWF, a high density of random cracks appeared in the network, leading to a decrease in the current pathways and therefore an increase in the resistance.

Because human vocalization is based on muscle movement and vibration, and because different vocalizations correspond to different muscle tensions or compressions, a new GWF sensor has been established to monitor throat muscle movements. This procedure is based on the relationship between the stress and resistance in a GWF, as opposed to the traditional method, which is based on the acoustic spectrum produced. When the assembled sensor is attached to the throat, GWFs are stretched or compressed along with the skin, thereby altering the resistance and collecting voice signals. The impressive sensitivity and electrical performance of GWFs allow for every English letter, along with certain Chinese characters, tones, and phrases, to be readily distinguished on the basis of their characteristic resistance changes and the characteristic peak of each vocalization. Additionally, the low sampling point in this study also simplified traditional speech recognition. Interestingly, we secured the crucial result that the signal waveforms nearly overlapped between vocalized and non-vocalized performances of the same speaking action.

2 Results and discussion

Figure 1(a) shows the key processes involved in fabricating and operating the GWF strain sensor. During the CVD growth of graphene [26], GWFs were formed by using a crisscross copper mesh [22]. During the transfer of GWFs, PDMS was used as an ideal adhesive on double-sided tape due to the flexibility, biocompatibility, high transparency, chemical inertness, and stability of PDMS over a wide range of temperatures [27]. Additionally, the double-sided tape was only 100 μm thick and much thinner than the previous medical tape [8], which led to more precise signals to distinguish different words from the tester. Though the strain sensor device was multi-layered, the total thickness was only about 200 μm, which was thin enough to obtain ideal signals and decreased attenuation. From the image of an integrated strain sensor in Fig. 1(c), it can be seen that the wearable device was rather thin and could be bent with excellent flexibility. As shown in Fig. S1 (in the Electronic Supplementary Material (ESM), this flexible strain sensor could fit human skin quite well, which resulted in a sizable strain from any direction. Figure 1(d) shows a scanning electron microscopy (SEM) image in which the wafers on the striped surface indicate that the GWF had a polycrystalline structure. If the assembled strain sensor was placed onto a human throat, signals of changing resistance could be obtained from the movement of throat muscle to monitor vocalization; this process is shown in Fig. 1(e). Furthermore, it was found that nearly the same signal waveforms were obtained when the tester performed the same action with or without vocalization, which could obviously aid patients who have difficulty in producing vocalizations.

In order to achieve enhanced performance in voice collection and recognition, the strain sensor should be able to distinguish every letter and the minimum structural unit of a word. Figure 2 shows test signal

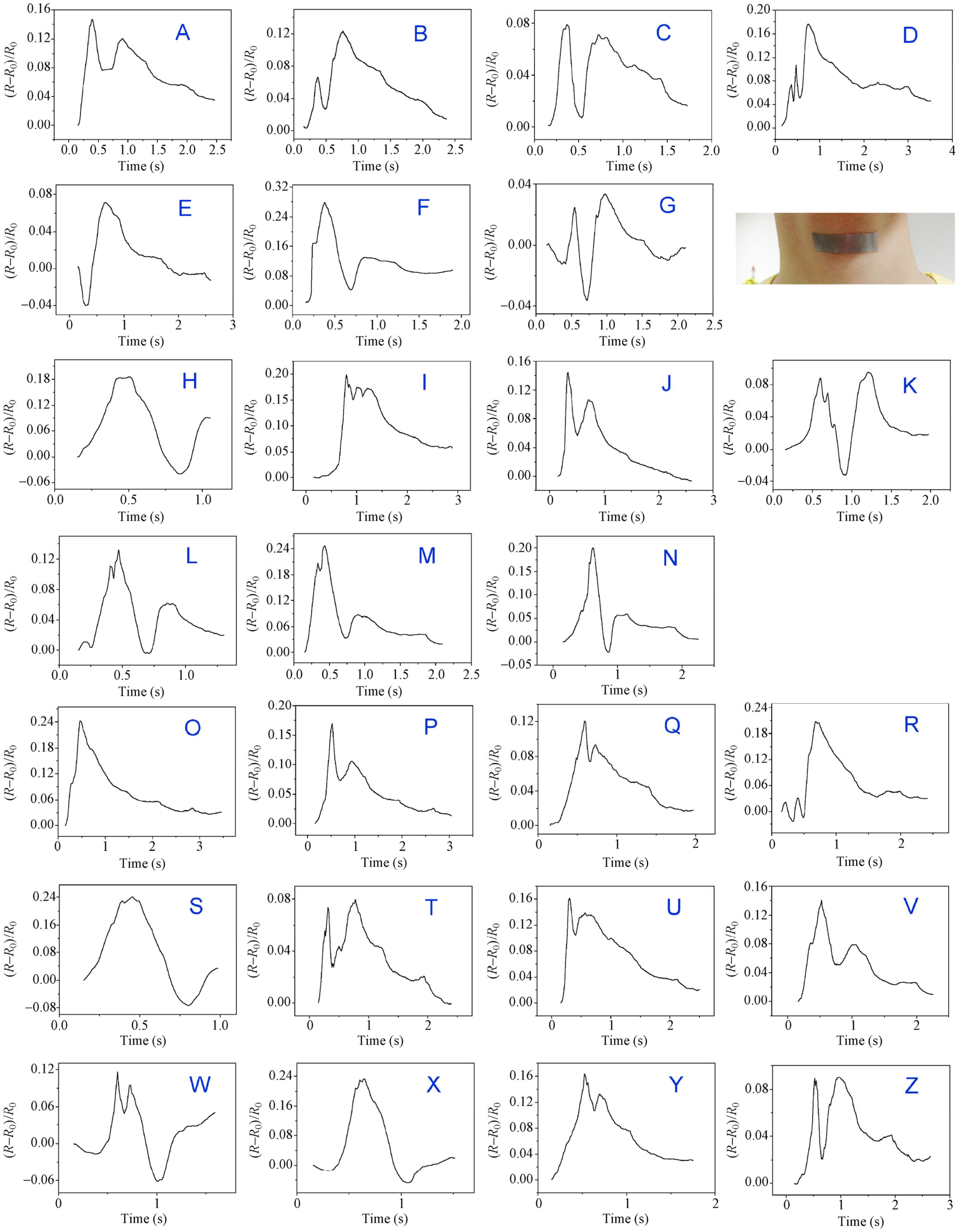

Figure 2 Recognition of 26 English letters.

waveforms of all 26 English letters; the inset is an actual test photograph. As expected, the waveforms are unique and repeatable for all letters. Every specific letter has its own different amplitude, duration, and resistivity variation. Since each individual speech organ is different, people can easily distinguish whether a given voice comes from the same person. Accordingly, we tested three females and a male speaking the same English letter, "A." As shown in Fig. S2(a) in the ESM, the key features of the test signal waveforms were similar. Placing sensors in the same spot each time on different people with different anatomy yielded very similar shapes of responses for the letter, whereas the same person repeating his/her own words generated relatively different patterns and magnitudes.

Louder sound requires more muscle movement, and thus the amplitude of the signal is expected to change with a change in volume. Compared with other methods of sound recognition, our sampling frequency was particularly low (fewer data points) compared to those from the method based on the produced sound spectrum. Nevertheless, one could still obtain the characteristic signal waveform of each letter, which simplified the sound collection compared with other traditional methods.

After clearly identifying each English letter, the voice recognition device was tested using different English phrases and sentences. As shown in Fig. 3, the resistance changed significantly when a tester said different words or phrases, such as "Graphene," "Lab of Science," "Strain Sensors," or "Graphene-based Strain Sensors." It was also found that each signal curve was apparently different (and the repeatability was quite impressive) by contrasting the same word ("Graphene") in Figs. 3(a) and 3(d) or the same phrase of ("Strain Sensors") in Figs. 3(c) and 3(d). Some other tests on monitoring English sentences are shown in Fig. S2(e) in the ESM. The signal proportion obtained from our multiple-strain sensors was at least 4% in resistance change, which was enough to measure with normal instruments (Keithley) and even a digital multimeter. Compared with ZnO (for which the maximal signal of current change was about 5 nA; voltage change was about 50 mV [28, 29]), CNTs (for which the maximal resistance change was about 2% [17, 30]), or molded graphene film-based sensors (for which the maximal current change was less than 0.8 μA [7, 31]), our GWF-based strain sensor demonstrated absolute predominance in voice recognition capabilities.

Different from English alphabets or phonetic

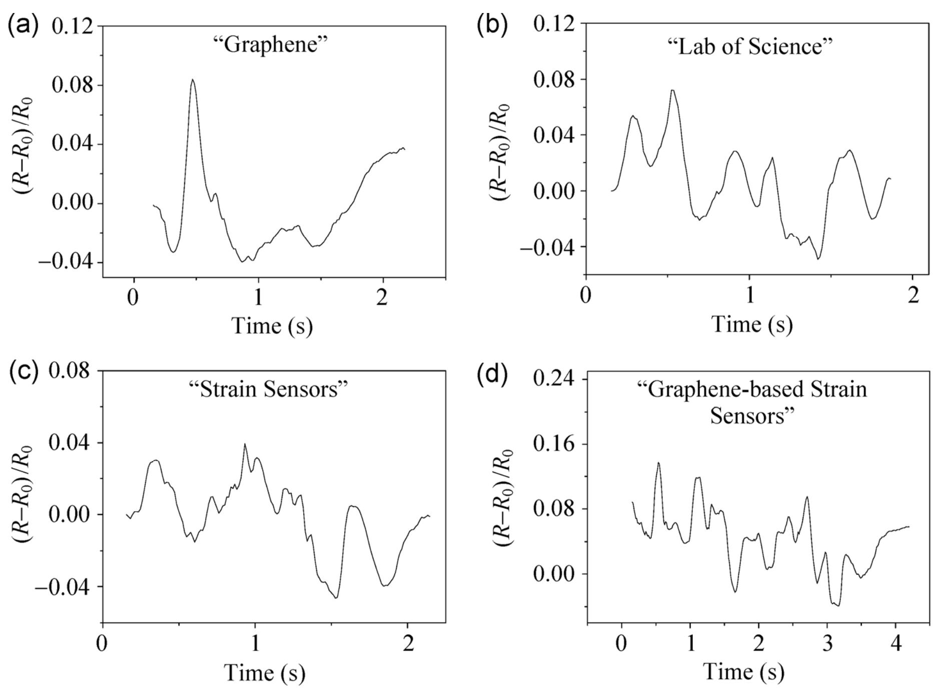

Figure 3 Recognition of English words and phrases. Relative resistance changes with throat muscle motions when the tester says (a) "Graphene," (b) "Lab of Science," (c) "Strain Sensors," and (d) "Graphene-based Strain Sensors."

symbols with both monosyllabic and polysyllabic words, Chinese characters are all monosyllabic, and encompass four pronunciation tones. Therefore, it is necessary to additionally identify Chinese characters and phrases with our GWF-based voice recognition sensors; the test results are shown in Fig. 4. In Figs. 4(a) and 4(b), respectively, the most frequently used Chinese characters of "东," "西," "南," and "北"—along with the Chinese tones of "ā," "á," "ǎ," and "à"—were tested. From the results, it was concluded that every Chinese character had its own characteristic peak, and that different Chinese tones generated different signals. Generally, signal waveforms of the third tone often had up and down, and signal amplitudes of the forth tone were always maximized. Another experiment concerning the characters "前," "后," "左," and "右"—along with the tones "ō," "ó," "ǒ," and "ò"—was performed (Figs. S2(b) and S2(c) in the ESM) in order to confirm the reliability of the data generated from our GWF-based voice recognition sensors. As shown in Figs. 4(c) and 4(d), Chinese phrases and sentences could also be recognized with our GWF-based sensors.

Because the collected signals for GWF-based sensors resulted from different degrees of stretching or shrinking strain, it is worth checking whether the signals of GWF-based sensors have the same response to loudspeakers, which rely on fluctuations in magnetic energy. Accordingly, audio experiments utilizing English and Chinese music with the GWF-based sensor on the vibrating membrane of a loudspeaker were performed; the results are shown in Fig. 5. It was found that the collected signals had a synchronous response to audio frequency, and could retain almost every characteristic peak. However, the volume of the loudspeaker also played an important role in the signal strength. As shown in Fig. S2(d) in the ESM, with an increase in the loudspeaker volume, the loudspeaker vibration amplitude was also increased, thereby increasing signal strength. The signals from the same sentence could be comprehensively obtained with the GWF-based sensor placed directly on the throat (red) and the loudspeaker (black), compared with the original recorded data (inset). Moreover, it can be seen that every characteristic peak is well recorded.

As mentioned previously, although the sampling frequencies of our GWF-based strain sensor were far lower than those of any existing recording software, this device could still maintain every sound's characteristic peaks with high fidelity. Figure 6 shows the

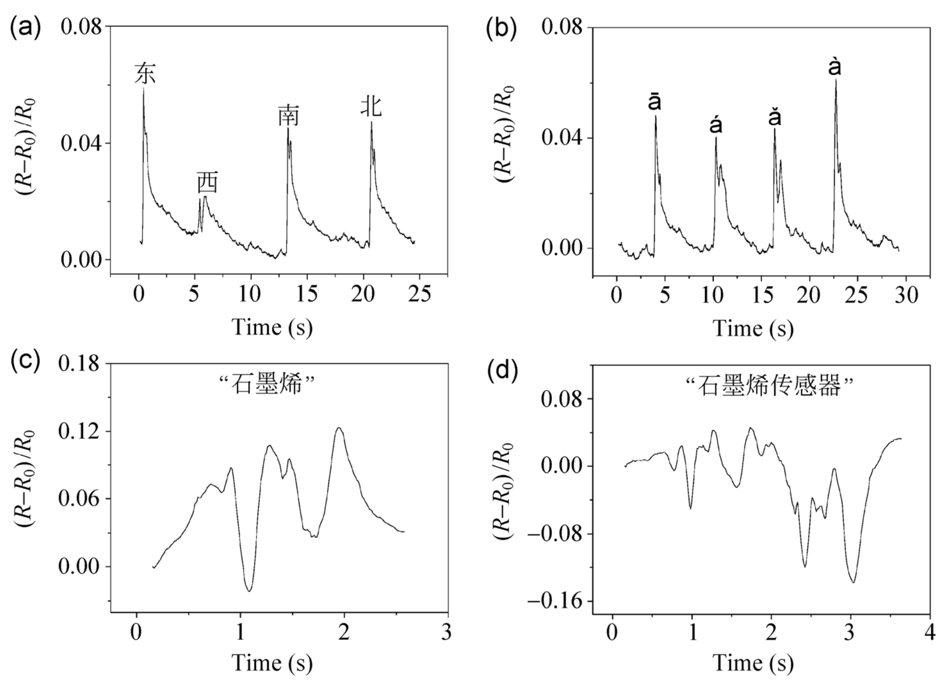

Figure 4 Recognition of Chinese characters, tones, and phrases. Relative resistance changes for (a) Chinese characters and (b) tones. Relative resistance changes with throat muscle motions when the tester says (c) "石墨烯" and (d) "石墨烯传感器."

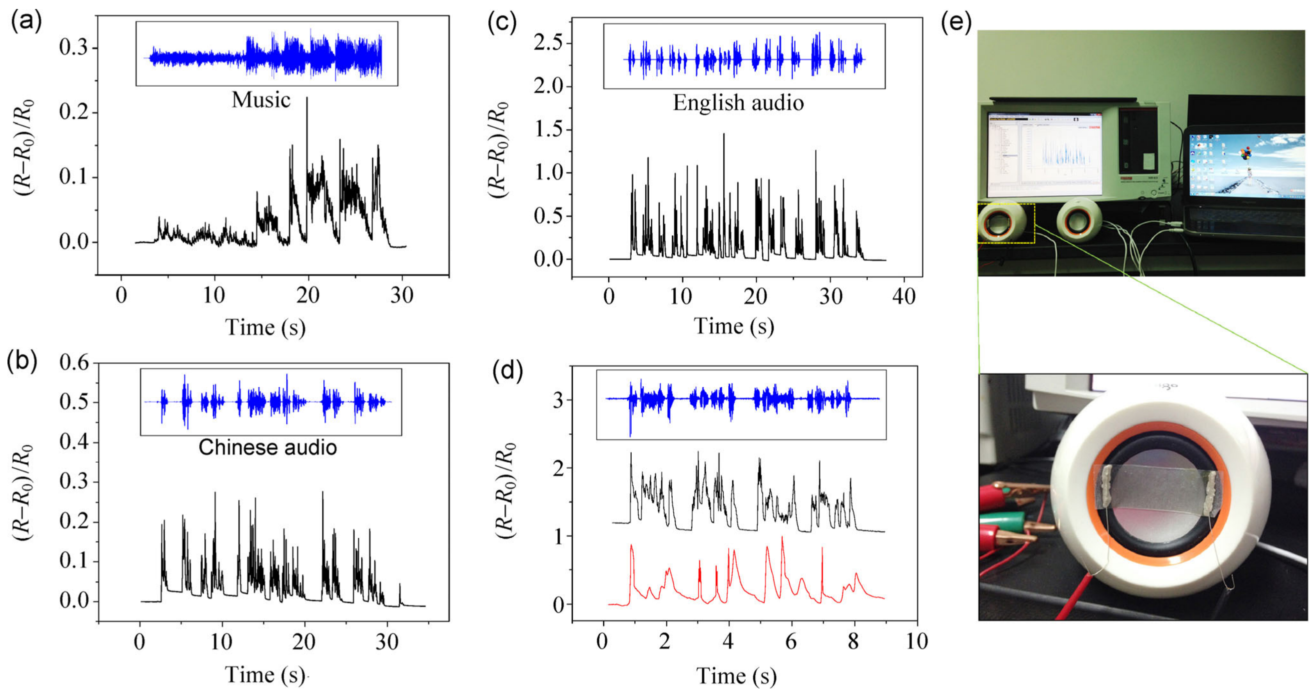

Figure 5 Recognition signals compared with the sounds from the loudspeaker. Relative changes of resistance when the loudspeaker played (a) music, (b) Chinese audio, and (c) English audio. (d) The same sentence tested by the sensors attached to the throat (red) and the loudspeaker (black). The insets show the original recording data. (e) Photograph of a strain sensor attached to the vibrating membrane of a loudspeaker.

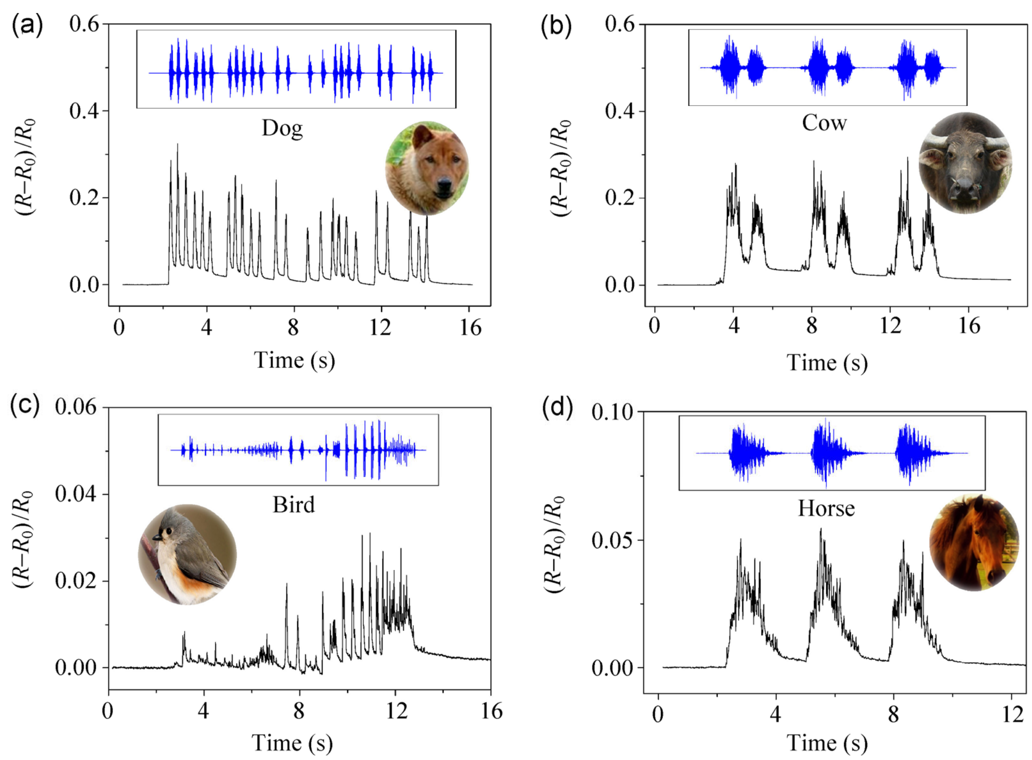

Figure 6 Recognition of animal sound recordings. Relative resistance changes when the loudspeaker played sound recordings of (a) a dog, (b) a cow, (c) a bird, and (d) a horse. The insets show the original recording data.

test data of four typical animal sounds (those of a dog, a cow, a bird, and a horse) and the insets display the original recorded audio data. Compared with the original recorded data, each measured signal from our GWF-based strain sensor had a high degree of consistency. Additionally, due to the excellent flexibility and high sensitivity, our GWF-based strain sensor might be used not only in the recognition of human sounds or the monitoring of the acoustic activity of animals, but also in the monitoring of various deformations or vibrations associated with robotics, fatigue detection, etc.

This work provides a first step in the proof of concept of the use of graphene sensors to detect different signals based on different tones and words. However, the results still require further validation, repetition, and cross-validation between different subjects, as well as an explanation regarding the origin of these signals (i.e., whether they are the result of muscle movement or a response to voice vibrations). In addition, signal processing should be performed through the mathematical extraction of signal shapes and features using automated mathematical algorithms, rather than the visual comparison of signal shape. For example, a fast Fourier transform (FFT) can be applied to both signals in order to yield the Pearson correlation coefficient, and to extract features of the given voice signal in order to check the similarity between two audio signals. A program based on the artificial neural network for comparing two audio signals has been developed to check the audio signal similarity, representing the first step towards the speech recognition (in the ESM). Furthermore, the utilization of machine learning algorithms on the extracted features could provide compelling proof that the different signals can indeed be differentiated and identified as letters and words.

3 Conclusions

In summary, a flexible and wearable acoustic detector was assembled by adhering GWFs on elastic polymer/double-sided tape film. The detector exhibited ultra-high sensitivity to tiny strains and vibrations, and could be used as an electronic skin covering the human throat to detect sound waves at low sampling frequencies. The results showed that the graphene sensor successfully collected and recognized all 26 English letters, some typical Chinese characters, phrases, and sentences. The graphene sensor could perfectly retain every signal characteristic of all peaks, which was highly consistent with the results from the loudspeaker experiment. Furthermore (and quite interestingly), it was found that nearly the same signal waveforms were obtained when the tester performed the same action with or without vocalization, which could aid patients who experience trouble in producing speech. Furthermore, because of its excellent flexibility, high sensitivity, good reversibility, and overall biocompatibility, the GWF-based sensor might become highly useful in earthquake monitoring, animal communication, and robotic voice development.

4 Materials and methods

4.1 Preparation of GWFs

Copper mesh (100 mesh, woven with wires 100 μm in diameter) was used as the CVD growing substrate. H_2 (~50 mL/min) and Ar (300 mL/min) flowed until a temperature of 1,000 °C was reached. At this stage, CH_4 (30 mL/min) was introduced into the reactor while the flow of H_2 was reduced to 5 mL/min and the flow of Ar was stopped to increase the number of graphene nucleation sites, because the GWF was more sensitive to deformation with a smaller grain. After 2 min, 200 mL/min of Ar was introduced for another 18 min. After GWFs were completely grown around the copper mesh, GWFs were obtained after the copper was etched away with an $FeCl_3$ and HCl aqueous solution.

4.2 Fabrication of GWF-based strain sensor

A small amount of PDMS (the weight ratio of base to cross linker was 10:1) was spin-coated on double-sided tape (100 μm thick). As-made materials were degassed under vacuum for 20 min to remove bubbles, and were then solidified at 80 °C for 1 h. A PDMS double-sided tape composite substrate (about 200 μm thick) was utilized. After GWFs were transferred to the PDMS-tape substrate, silver wires were connected with silver paste, and the multilayer device was assembled.

4.3 Custom-made data processing system

The surface of the GWF was observed with an optical microscope (Axio Scope A1) and a scanning electron microscope (LEO 1530). Audio tests of the strain sensors were performed with a computer-controlled loudspeaker. Voice collection and recognition of the GWF-based sensor were performed with a digital meter (Keithley 4200-SCS) with a test step of 5 ms and a source-drain voltage of 1 V DC.

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Electronic Supplementary Material: Supplementary material (photographs of GWF strain sensors adhered on human skin with deformation, and supplementary recognition signals for different people and volumes, Chinese characters, tones, and sentences) is available in the online version of this article at http://dx.doi.org/10.1007/s12274-014-0652-3.

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