Detecting Subtle Vibrations Using Graphene-based Cellular Elastomers

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ABSTRACT

Ultralight graphene elastomer-based flexible sensors are developed to detect subtle vibrations within a broad frequency range. The same device can be employed as an accelerometer, tested within the experimental bandwidth of 20-300 Hz as well as a microphone, monitoring sound pressures from 300 to 20,000 Hz. The sensing element does not contain any metal parts, making them undetectable by external sources and can provide a sensitivity of 2.6 mV/g, which is higher than those of rigid Si-based piezoresistive micro electro mechanical systems (MEMS).

Detection of subtle vibrations is important for a wide range of applications including rehabilitation ¹, activity recognition ², and biomechanical tracking ³, all of which require compliant wearable sensory parts. Conventional micro electro mechanical systems (MEMS) consisting of accelerometers, gyroscopes and microphones were proposed to detect small changes in displacement, velocity and accelerations for body motion analysis ⁴⁻⁵, fall detection ⁶, to measure respiratory rates⁷ and perform electrocardiogram⁸, to name few. While these well-refined devices can offer high sensitivity within a broad bandwidth, they require complex fabrication processes performed in sophisticated clean-room facilities and often complex circuitries. Additionally, traditional MEMS devices consist of rigid structures, which are unable to truly fulfil the requirements in emerging flexible electronics applications.

In recent years numerous flexible sensing elements using graphene, carbon nanotubes and metallic nanoparticles embedded in polymeric/elastomeric materials have been proposed as alternatives to MEMS based sensors. While such alternatives are suitable for quasi-static detection of classical mechanical quantities such as, strain ⁹⁻¹², force ¹³ and pressure ¹⁴⁻¹⁶, they have very limited response to detect vibrations, underperforming even in comparison to human skin (essentially a viscoelastic tactile sensor) which can comfortably detect dynamic excitations up to hundreds of Hertz ¹⁷⁻¹⁸. The polymer elastomers employed in these sensors have relatively large elastic moduli and a poor response to high frequency excitations due to their viscoelasticity and poor mechanical recovery ¹⁹⁻²¹. Although engineering of the architecture of the sensing components can potentially improve the detection sensitivity, the dynamic performance of previously reported polymer elastomer-based flexible sensors are still not comparable to those of conventional Si-based MEMS devices.

Recently, we ²²⁻²³ and other groups ²⁴⁻²⁹ have fabricated graphene-based elastomers (g. elastomers) by using graphene as a building block. This new class of graphene-based

elastomeric material possesses a range unique features that are unattainable in conventional polymer elastomers. For example, graphene elastomers are extremely soft, their Young's modulus are 5,000~1,000,000 times lower than that of PDMS (a widely used polymer elastomer for flexible electronics); they are inherently piezoresistive and possess excellent response to dynamic force²²⁻²³. These unique features make graphene elastomer a promising candidate for sensing vibrations.

In this work, we investigate the vibration sensing capability of graphene elastomer, and demonstrate that the graphene elastomer can be used in sensing small vibrations with frequencies ranging from 20 to 20,000 Hz, including uniaxial inertial loads (20-300 Hz) and sound waves within the audible frequency range (300 to 20,000 Hz). Their sensitivities are much higher than those of polymer elastomer-based sensors and even comparable to or better than the sophisticated Si-based piezoresistive MEMS devices³⁰⁻⁴⁰.

The devices were fabricated through previously developed freeze casting method²³. (Details of the sample preparation and experimental procedures are provided in Supporting Information Section 1). The resultant cylinder shaped graphene elastomer (with 10 mm height and 12 mm diameter) revealed a honeycomb-like cellular structure and an ability to recover from large deformation (Figure 1 (a-b)). Interestingly, mechanical properties of the graphene elastomers can be tuned through the modification of density during the fabrication by simply altering the concentration of the graphene oxide dispersion. Our previous finding indicates that higher density graphene elastomer showed better dynamic response than lower density graphene elastomer²². (see Supporting Information Section 2 for the details.) Therefore, we focus on the sensing capabilities of a relatively dense graphene elastomer with 5 mg/cm³ density for the high frequency applications. Results for a low density graphene elastomer-based device have also been also provided for comparison (Figure S1).

Figure 1 – (a) A Scanning Electron Microscope (SEM) Image showing the cellular structure of the graphene elastomer (g. elastomer) with a 5 mg/cm^3 density. (b) Force-compression curve for 5 mg/cm^3 device up to 10% compression. Inset shows the compression-release curve up to 80% where the device exhibits elastomeric behaviour.

To demonstrate the vibration sensing capability, a cylindrical proof mass was placed on top of the graphene elastomer-based sensor device to convert the vibration to the form of acceleration. As shown in Figure 2 (a), the device was excited with a sinusoidal loading generated by a commercial shaker with frequencies ranging from 20 to 300 Hz (limited by the reference commercial accelerometer's operation bandwidth). Figures 2 (b-d) show the response of the device under 40, 100 and 240 Hz actuation in the time domain. All three cases provided stable and consistent output and tracked the reference accelerometer well. As shown in Figure 2 (b), the phase difference between the device output and the commercial accelerometer is more pronounced at 40 kHz, which is within close proximity of the resonant frequency (see detailed discussion on the resonant frequency of the graphene elastomer in Supporting Information Section 4).

Figure 2 –Schematic of the test setup for accelerometer experiments. (a) Voltage output of the graphene elastomer-based sensor (G. Elastomer) under sinusoidal excitation at (b) 40 Hz, (c) 100 H with 1_{g_{pk-pk}} and (d) 240 Hz with 6 _{g_{pk-pk}} acceleration.

The graphene elastomer-based device can successfully measure up to 6g peak-to-peak acceleration and exhibits a linear response to the changes in the amplitude of the acceleration when the acceleration is varied from 1 _{g_{pk-pk}} to 6 _{g_{pk-pk}} at several frequencies (Figure 3 (a)). Having known the peak-to-peak voltage output of the graphene elastomer for the corresponding acceleration, the acceleration sensitivity of the graphene elastomer is calculated for various excitation frequencies from the experiments where the device is excited by an acceleration of

1 g_{pk-pk} (Figure 3 (b)). Results show a high sensitivity at lower frequencies, which is due to low stiffness/proof mass ratio. This is also consistent with our theoretical approximation which indicates a resonance at 33 Hz considering the load-deformation curve in Figure 1 (b) (details available in the Supporting Information Section 4). The device sensitivity (S) reaches an average value of 2.6 mV/g above 150 Hz which is up to an order of magnitude higher without signal conditioning when compared to recent well-refined MEMS-based piezoresistive accelerometers³⁰⁻⁴⁰.

Apart from its excellent sensitivity, the graphene elastomer sensor also revealed excellent device performance, including good tracking comparability to the commercialized accelerometer and low noise level. The degree of linearity between the output signals of graphene elastomer device and the reference accelerometer was evaluated through a coherence analysis commonly used for accelerometer characterisation, and the result shows good correlation between graphene elastomer sensor and commercialized accelerometer^{34, 41} (Figure S4). Additionally, to calculate the acceleration resolution of the device, its noise floor was analysed (Figure S5) to find the minimum detectable voltage bounded by the thermal noise. Utilizing the measured thermal noise floor for the device (10.36 $\mu\text{V}/\sqrt{\text{Hz}}$) and the corresponding device sensitivity (Figure 3 (b)), we have found that the graphene elastomer-based sensor devices can detect accelerations as small as 10 mg (Figure S6). Importantly, all the data presented here was not filtered or amplified with the circuitry and raw output is presented in order to capture the intrinsic performance of the device. These results indicate excellent performance of the graphene elastomer-based accelerometer.

Figure 3 - (a) Voltage output (shown in logarithmic scale) of the graphene elastomer (G. Elastomer) ($5\text{mg}/\text{cm}^3$) at different acceleration amplitudes and frequencies with an absolute error of ± 0.527 mV. (b) Sensitivity of the device with respect to vibration frequency obtained from $1\text{g}_{\text{pk-pk}}$ experiments. Absolute error originating from the measurement equipment is ± 1.25 mV/g at 40 Hz and less than ± 0.6 mV/g above 100Hz. Please see the Supporting Information Section 5 for the details regarding the measurement errors.

To further investigate the vibration sensing capability of the graphene elastomer at higher frequencies, the device was tested under sound pressures in the audible frequency range (up to 20 kHz) generated by a speaker positioned in the proximity, hence demonstrating its use as a microphone. For these experiments, we have again employed the same graphene elastomer-based device with identical density in the same configuration, but without the proof mass as the nature of the sound pressure is not inertial. The device can successfully track the reference microphone output when a constant amplitude sinusoidal sound wave is swept within the bandwidth of the speaker, 300-20,000 Hz (Figure 4 (b) and Supplementary Video 1). More interestingly, the device can successfully track sound waves applied simultaneously at different frequencies (Supplementary Video 2). Figure 4 (b) demonstrates the power spectrum of the graphene elastomer when excited with single tone sinusoidal sound waves with various frequencies resulting in a clear trend from 1 to 20 kHz (the upper frequency limit defined by

the bandwidth of the speaker used in the experiments). Moreover, we observe that graphene elastomer can successfully respond to the changes in the amplitude of sound waves, which have been varied at the same frequency (Figure 4 (c)). Although the sensor does not possess an optimized geometry for the microphone applications, it shows good dynamic response to sound waves, indicating promising performance in microphone application.

Figure 4 – (a) A schematic of the test setup for microphone application. (b) Frequency response to sound pressures from 300 Hz to 20 kHz. The amplitude of the sound wave was kept constant and the gain of the speaker was set as 100% for this experiment. (c) Changes in the peak amplitudes of the power with respect to the variations in sound amplitude (varied by speaker gain) at various frequencies. Standard deviation based on 3 measurements have been presented in the form of error bars separately for each data points. Overall the relative error ranges from $\pm 2\%$ to $\pm 8\%$.

The excellent vibration sensing capability of the graphene elastomer originates from its ultralow modulus and fast response nature. Owing to the ultralow modulus (ultralow stiffness) nature of the graphene elastomer, the subtle pressures induced by the inertial loading or the sound waves can result in sufficiently large deformations, leading to measureable changes in electrical resistance. Additionally, the stress transmission rate of graphene elastomer is comparable to those of dense polymer elastomers, enabling the fast response to high frequency dynamic vibration. Combining these unique features, graphene elastomer-based sensor can detect subtle vibration with a broad range frequency and provide outstanding performance in the application of accelerometer and microphone

In conclusion, we developed a soft vibration sensor based on the graphene elastomer, and further demonstrated its potential applications as an accelerometer and a microphone. The graphene elastomer-based accelerometer provides higher sensitivity and better synergy with wearable sensing applications than those of conventional rigid piezoresistive MEMS devices. Additionally, the graphene elastomer-based microphone is able to detect sound waves with frequency range of 300 - 20,000 Hz. Noteworthy, the excellent vibration sensing capability is rarely found in flexible electronics made from the conventional polymer elastomers, as their viscoelastic behaviour and relatively high modulus limit their responsive behavior to subtle vibrations. This new class of soft vibration sensor is promising for novel flexible electronics applications, soft robotics and wearable sensing technologies. Finally, the devices contain practically no metallic parts, which prevents them from being discovered through external sources. This has a significant potential to be used in a number of systems requiring specialised undetectable sensing units (useful for instance in defence related applications).

ASSOCIATED CONTENT

Supporting information for the following content has been included: experimental procedures; effect of density on the device sensitivity under dynamic loadings; cross-sample comparison: vibration response of two graphene elastomers with same density; theoretical estimation of the resonant frequency; standard deviation of the peak-to-peak voltage measurements and sensitivity; coherence analysis; thermal noise measurements; resolution of the accelerometer; readout circuit for the accelerometer application; readout circuit for the microphone application. Supporting Information (ESI) available free of charge via the Internet at <http://pubs.acs.org>.”

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Abstract Figure

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