Graphene “Microdrums” on Freestanding Perforated Thin Membrane for High Sensitivity MEMS Pressure Sensor

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Abstract

We present a microelectromechanical systems (MEMS) graphene-based pressure sensor realized by transferring a large area, few-layered graphene on a suspended silicon nitride thin membrane perforated by a periodic array of micro-through-holes. Each through-hole is covered by a circular drum-like graphene layer, namely graphene “microdrum”. The introduction of the through-holes into the supporting nitride membrane allows generating an increased strain in the graphene membrane over the through-hole array by local deformations of the holes under an applied differential pressure. Further reasons contributing to increased strain in the devised sensitive membrane include larger deflection of the membrane than that of its imperforated counterpart membrane, and direct bulging of the graphene microdrum under an applied pressure. Electromechanical measurements show a gauge factor of 4.4 for the graphene membrane and a sensitivity of $2.8 \times 10^5 \text{ mbar}^{-1}$ for the pressure sensor with a good linearity over a wide pressure range. The present sensor outperforms most existing MEMS-based small footprint pressure sensors using graphene, silicon, and carbon nanotubes as sensitive materials, due to its high sensitivity.

Keywords: Graphene; MEMS; pressure sensor

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Graphene is a promising material for applications in microelectromechanical systems (MEMS) owing to its atomic thickness, fast electron mobility,\textsuperscript{1,2} and high Young’s modulus.\textsuperscript{3-5} Because a single layer of graphene is impermeable to standard gases including helium\textsuperscript{6,7} and has strong adhesion to silicon oxide (SiO\textsubscript{2}) substrate,\textsuperscript{8} graphene has been suggested as an atomic thick pressure sensor,\textsuperscript{7} a separation barrier between two distinct regions,\textsuperscript{9,10} and a high-performance drumhead resonator.\textsuperscript{11} Recently, chemical vapor deposition (CVD) has enabled large-area uniform formation of single and few-layer graphene sheets on different substrates.\textsuperscript{12-14} This ability, in conjunction with well-developed patterning and transferring methods for graphene sheets,\textsuperscript{15-23} have opened up new opportunities of developing graphene-based sensors and actuators. Strain induced electrical-mechanical coupling in graphene are widely reported.\textsuperscript{17,24-29} At present a few MEMS-based graphene pressure sensors have been demonstrated.\textsuperscript{7,30-32} In a pioneering work on graphene pressure sensors, a graphene membrane was suspended over a shallow well etched into a SiO\textsubscript{2} layer grown on a silicon substrate, where the piezo resistive effect provided direct electrical readout of pressure to strain transduction and was demonstrated to be independent of crystallographic orientation.\textsuperscript{7} Another remarkable pressure sensor design involved forming a graphene membrane on a silicon nitride (SiN\textsubscript{x}) membrane suspended over a micromachined silicon base.\textsuperscript{30,31} Also, a different pressure transducer was developed by using graphene flakes to cover an array of wells engraved into a fixed SiO\textsubscript{2} layer grown on a silicon substrate.\textsuperscript{32} The aforementioned graphene-based MEMS pressure sensors have a compact footprint of sub-mm\textsuperscript{2} and even smaller. In another category of graphene-based pressure sensors, large area graphene-polymer composite and laser-scribed graphene foam have been used as sensitive materials.\textsuperscript{33,34} These sensors provided tremendous sensitivity, but had a large sensing area on the order of square centimeters and even larger.

In this paper, we report on a high sensitivity, small area MEMS pressure sensor using few-layered graphene on a flexible perforated SiN\textsubscript{x} thin membrane (Figure 1(a, b)). The SiN\textsubscript{x} membrane acts as a supporting layer for the graphene membrane and has a periodic array of microsized through-holes (Figure
Therefore, an array of circular drum-like graphene structures, namely graphene “microdrums”, are formed above these through-holes (Figure 1(e)). Compared to the previously reported sensor designs of using a standalone graphene membrane and an imperforated nitride-graphene composite membrane as sensing elements, the introduction of the microsized through-hole array into the supporting membrane allows generating an increased membrane strain locally in the graphene layer over the holes (Figure 1(c)). Further reasons which add to obtain a large strain change in graphene and thus a high pressure sensitivity of the sensor include the facts that the perforated membrane deflects more than an imperforated counterpart membrane of the same dimensions, and that the graphene microdrums are pressurized to bulge up under an applied pressure.

To proof this new device concept, we fabricated a perforated SiN₃ square membrane (490 x 490 µm²) by depositing 200±2.7 nm thick nitride on a silicon substrate and patterning with 2.5 µm-diameter holes, followed by removing silicon below the membrane. Subsequently, a few-layered graphene membrane (~2 nm thick or ~6 atomic layers) was transferred on the perforated nitride membrane. The nitride membrane was pretreated with oxygen plasma to improve van der Waals interactions between the graphene and nitride membrane. After that, the graphene resistor pattern was patterned with the help of a shadow mask. Lastly, metal contacts were formed by using shadow mask evaporation of gold. See Methods section for details of device fabrication. To test the fabricated device, the backside of the device was adhered to the outlet of a plexiglass-based air channel. Air pressure was applied from the inlet of the air channel using a programmable syringe pump. A commercial differential pressure sensor was used to measure differential pressures applied across the sensitive membrane. See Methods section for details of the testing setup.
Figure 1(d) shows the surface coverage of graphene on the perforated nitride membrane suspended over the micromachined silicon base. Only a few pinholes were observed in the graphene membrane (see arrows in Figure 1(e)), which may be introduced during the graphene deposition and/or the transfer process. To confirm that the graphene membrane stayed bonded with the nitride membrane within a range of applied pressures, we performed contact profile measurement (Figure 1(f)). Figure 1(g) shows that the measured maximum deflection of the composite membrane is 14.1 µm at a differential pressure of 400 mbar. Let us assume that the pressurized graphene is totally detached from the supporting membrane. Then, according to mechanical simulations, the maximum deflection of 46 µm will be expected at the center of the membrane, which is much larger than the measured deflection mentioned above. Therefore, it was likely that the graphene adhered well with the nitride membrane. In fact, no detachment of the graphene from the perforated nitride membrane was observed even when the membrane popped out under the air pressure of ~600 mbar.
Figure 1. (a) Schematic of the proposed MEMS pressure sensor using a graphene membrane on a perforated SiNₓ thin membrane formed on a micromachined silicon base. (b) Optical image of the fabricated pressure sensor. (c) Simulated deformation of the membrane and shape distortion of the through-holes. (d, e) SEM images of the graphene membrane on the perforated SiNₓ membrane. The white arrows in (e) indicate the locations of some pinholes in the graphene. The inset of (e) shows the standalone circular graphene microdrums. (f) Optical images of the sensor before and after applying a differential pressure of 400 mbar. (g) Measured surface profile of the graphene-perforated SiNₓ composite membrane along the line A-A’ across the center of the membrane. The measurement was conducted using Ambios XP-100 Stylus contact surface profiler.
The piezoresistive effect of the graphene sensor was measured with a Wheatstone bridge circuit (Figure 2(a)). A small input voltage of 20 mV was applied across the junctions of two shunt resistive circuits. The total resistance of the graphene sensor \( R_{\text{tot}} \) is composed of \( R_{g} \) of the graphene on the suspended square membrane, \( R_{g1}, R_{g2}, R_{g3} \) and \( R_{g4} \) of graphene in the surrounding regions, and the contact resistance \( R_{c} \) between the metal contacts and graphene. \( R_{\text{tot}} \) was measured to be 1215 Ω. The relative resistance change of the sensor \( \Delta R_{\text{tot}}/R_{\text{tot}} \) can be related to the output and input voltages \( (V_{\text{out}} \) and \( V_{\text{in}} \)) of the sensor by eq 1:

\[
V_{\text{out}} = V_{\text{in}} \left( \frac{R_{3}}{R_{3}+R_{\text{tot}}} - \frac{R_{2}}{R_{1}+R_{2}} \right)
\]  \hspace{1cm} (1)

where \( R_{1} \) and \( R_{2} \) were chosen to be the same and \( R_{3} \) was adjusted till a balanced bridge circuit was obtained. The output voltage variation is quasi-linearly proportional to \( \Delta R_{\text{tot}} \) and described as:

\[
\frac{V_{\text{out}}}{V_{\text{in}}} \approx \frac{\Delta R_{\text{tot}}}{4R_{\text{tot}}}
\]  \hspace{1cm} (2)

Based on the equivalent circuit of the sensor shown in Figure 4(a), \( R_{\text{tot}} \) is expressed in eq 3:

\[
R_{\text{tot}} = R_{g1} + \frac{1}{\frac{1}{R_{g3}} + \frac{1}{R_{g}} + \frac{1}{R_{g2}}} + R_{g2} + 2R_{c}
\]  \hspace{1cm} (3)

As a result, the relationship between \( \Delta R_{\text{tot}} \) and \( \Delta R_{g} \) is obtained in eq 4:

\[
\Delta R_{\text{tot}} \approx \frac{\Delta R_{g}}{(1+\frac{R_{g}}{R_{g3}}+\frac{R_{g}}{R_{g4}})^2}
\]  \hspace{1cm} (4)

Based on the dimensions of the resistors in Figure 2(a), the graphene on the suspended square membrane is estimated to be \( R_{g} = 1473 \) Ω. The relative resistance change of this part of the graphene can be written as \( \Delta R_{g}/R_{g} = 3.6 \Delta R_{\text{tot}}/R_{\text{tot}} \). As \( R_{g3} \) and \( R_{g4} \) are in parallel with \( R_{g} \), their values can largely influence the measured electrical signal. If \( R_{g3} \) and \( R_{g4} \) are too low, the output voltage signal will be greatly suppressed. Therefore, the graphene in the areas other than those labeled in Figure 2(a) were removed during the device fabrication.
Results and discussion

Figure 2(b) shows the output voltage normalized to the input voltage of the device responding to an increase in step-like differential pressure. The output voltage rose with increasing air pressure applied to the graphene-perforated membrane. At a differential pressure of 350 mbar, 0.067% relative change was observed at the output voltage, corresponding to 0.97% change in the resistance. The rapid rise of the output signal indicates an immediate piezo-resistive response to the pressure applied to the membrane. Based on the noise floor of the output signal shown in Figure 2(b), the noise equivalent pressure resolution of the sensor is about 30 mbar, which can be further improved by optimizing the detection circuit, e.g., using a low-pass filter and a low-noise amplifier.

![Diagram](image)

**Figure 2.** (a) Schematic of the equivalent circuit of the graphene sensing element connected into a Wheatstone bridge circuit. (b) Voltage response of the device to step like increasing differential pressures. (c) Voltage response of the device to rapid increase and gradual decrease in applied differential pressure.
Figures 2(d) show the results of cyclic pressure testing for the device. The experiment involved rapidly applying differential air pressure to the sensitive membrane by pumping air and then gradually releasing the pressure. In Figure 2(c), the pressure pump time was controlled from 7 s to 1.5 s while the pressure release time was decreased from 18 s to 2 s by adjusting the air pumping and withdrawal speed of the pressure control apparatus. It is clear that upon applying an air pressure, the output voltage was able to quickly follow the sudden increase of the internal pressure and then go back to the baseline. The response time here is mainly determined by the pump and vent speed, so the actual response time is expected to be faster.

Figure 3 shows the static response of $V_{out}/V_{in}$ of the device with respect to applied pressure. A good linearity was observed and the sensitivity of $3.88 \times 10^{-5}$ mV/mbar was obtained. The gauge factor $G$ of graphene for the sensor was estimated by $G = \frac{\Delta R}{R} / \frac{\Delta L}{L} = 4.4$ at 350 mbar. Here, the average strain of the suspended square membrane was calculated to be 0.22% for 14.3 µm deflection at the center of the membrane. The obtained gauge factor of the graphene used here is comparable to other reported CVD-grown graphene. For example, the reported gauge factor is 2.92 for the standalone graphene, 6.1 for the graphene on poly(dimethylsiloxane) substrate, and 1.6 for the graphene on the SiN$_x$ membrane.
We studied the roles of the perforated SiN membrane and the graphene microdrums over the through-holes in determining the sensitivity of the sensor. First, mechanical responses of the graphene coated perforated nitride membrane to different applied pressures were visualized using a 3D optical surface profiler (ZYGO Newview, Middlefield, CT). As shown in Figure 4(a), under 415 mbar differential pressure, the membrane was deformed into a convex shape with a maximum out-of-plane deflection of 14.3 µm at its center. The measured surface profiles of the membrane under other differential pressures were also given in Figure 4(b). For a square imperforated nitride membrane, the maximum out-of-plane deflection δ with respect to differential pressure P can be described with the following equation:\(^\text{39, 40}\)

\[
P = \frac{B_1 \sigma_0}{(a/2)^2} \delta + \frac{B_2 f(v) t E}{(a/2)^4 (1-v)} \delta^3 \tag{5}
\]

where \(B_1\) and \(B_2\) are dimensionless constants, \(\sigma_0\) is the initial stress, \(E\) is Young’s modulus, \(a\) is the side length of the square membrane, \(v\) is the Poisson ratio, \(f(v)\) is a geometry function, and \(t\) is the thickness of the membrane. \(B_1 = 3.45, B_2 = 1.994, v = 0.22,\) and \(E = 239\) GPa were taken from Refs [40, 41]. Previous research shows that perforated membrane can be replaced with imperforated one with modified elastic modulus in the numerical calculation.\(^\text{42}\) Thus, the eq 5 can also be applied to imperforated membrane.
Figure 4(c) shows the fitted results for the graphene coated perforated membrane, as well as the graphene coated imperforated counterpart membrane with the same dimensions for comparison purpose. It was obtained that $\sigma_0=58$ MPa and $f(\nu)=0.32$ for the perforated membrane, while $\sigma_0=41$ MPa and $f(\nu)=0.65$ for the imperforated one. By using the obtained deflection equations for both of the perforated and imperforated membrane, the ratio of maximum deflection between the perforated and imperforated membrane can be expressed by eq 6:

$$\frac{d_{\text{perforated}}}{d_{\text{imperforated}}} = 0.533 \times P^{0.08} \quad (6)$$

Under the differential pressure of 415 mbar, the imperforated membrane with graphene had the maximum deflection of 11.7 $\mu$m, which was 2.6 $\mu$m less than that the perforated membrane with graphene. The eq 6 also indicates that further increasing differential pressure will not significantly improve the deflection of the graphene coated perforated membrane compared to the imperforated counterpart membrane, and therefore, will have some but limited effect to improve pressure sensitivity of the device.
Figure 4. (a) Measured 3D surface profile of the graphene coated perforated nitride membrane under 415 mbar differential pressure. (b) Deflection profiles of the membrane across the middle line of the perforated membrane (parallel to the side of the membrane) under various differential pressures. (c) Maximum deflection at the center of the membrane as a function of differential pressure for the perforated and the imperforated membrane. The black and red dots are the experimental data. The black and red lines are the fitted curves obtained using eq 5.

Next, we conducted mechanical simulations to illustrate strain distributions in both of the perforated and imperforated SiNₓ membranes, each including the ~2 nm thick graphene layer. The simulations were carried out through finite element method based commercial package (COMSOL Multiphysics). Limited by computational power, a reduced model of side length 200 µm was calculated for the purpose of illuminating the working mechanism. Under 500 mbar differential pressure, the imperforated membrane had the maximum areal strain of 0.14% at the center of the membrane with the
deflection of 3.49 µm (Figure 5(a)). For the perforated membrane, a similar strain distribution was observed. In the non-hole areas of the membrane the maximum areal strain was found to be 0.15%, which was only slightly higher than that observed in the imperforated counterpart membrane. However, the maximum areal strain in the graphene layer over the holes reached 0.34% at the center of the membrane with the maximum deflection of 4.13 µm. Therefore, the maximum strain in the hole areas was as high as 2.27 times that occurred in the non-hole areas of the perforated membrane. Furthermore, the average areal strain along the line across the center of the perforated and imperforated membranes was 0.203% and 0.12%, respectively (Figure 5(c)). Although the maximum deflection of the perforated and imperforated membranes differed only by 18.3% (3.49 µm vs. 4.13 µm), the average strain in the perforated membrane increases by 62.4% (0.203 % vs. 0.12 %) due to the introduction of the through-holes into the SiNx membrane.

Furthermore, as the differential pressure was applied to the graphene coated perforated membrane, the graphene microdrums over the holes also bulged into a curved shape. To illustrate how this bulging factor affected the pressure sensitivity of the device compared to the in-plane membrane strain, let us imagine a state when the strain in the pressurized SiNx membrane is suppressed, i.e., the holes stay in the plane and maintain the original circular shape with a diameter of 2.5 µm. Simulations showed that, under 500 mbar differential pressure, the graphene microdrum will deflect by 9.1 nm at its center and the average strain of 0.0035% will be obtained over the whole microdrum. The magnitude of this strain is about two orders of magnitude lower than the aforementioned maximum strain of 0.34% in the microdrum. Therefore, the bulging of the pressurized circular graphene had a limited influence on the overall strain change of the microdrum. As a matter of fact, the previously reported graphene-based pressure sensors employed the bulging effect of the graphene suspended over the wells in the fixed substrate, thus offering relatively low sensitivity.\(^3\)\(^2\) Comparison between the effects of membrane strain
and bulging, it is evident that the inhomogeneous membrane strain of the perforated membrane was the key to the improved pressure sensitivity of the device.

![Figure 5](image)

**Figure 5.** (a) Simulated areal strain under a differential pressure of 500 mbar for the imperforated (left) and perforated (right) membranes. The z coordinate and the color scale show the amplitude of the areal strain. (b) The areal strain along the line across the center of the perforated and imperforated membranes.

Table 1 compares our device with the recently reported graphene-based MEMS/NEMS pressure sensors. Generally, the sensitivity of piezoresistive pressure sensors can be calculated using \( S = \frac{\Delta R}{R \times P} \). Our sensor has the sensitivity of \( 2.8 \times 10^{-5} \) mbar\(^{-1} \) which outperforms most of the reported graphene, silicon,
and carbon nanotubes based MEMS/NEMS pressure sensors. Specifically, the present sensitivity is higher than 2.96×10^{-6} mbar^{-1} of the standalone graphene membrane-based sensor and 6.67×10^{-6} mbar^{-1} of the sensor using the graphene meander patterns on imperforated SiNx membrane. As mentioned above, another previous pressure sensor used a graphene membrane suspended over the wells made in a SiO2 layer on the bulk silicon substrate, where the resistance variation only came from the bugling effect of the graphene. The resulting sensitivity of that sensor was about 32 times of magnitude lower compared to our sensor.

**Table 1** Performance comparison among MEMS pressure sensors

<table>
<thead>
<tr>
<th>Device structure</th>
<th>Dimensions (µm²)</th>
<th>Sensitivity (mbar⁻¹)</th>
<th>Ref</th>
</tr>
</thead>
<tbody>
<tr>
<td>Graphene on suspended perforated membrane</td>
<td>490×490</td>
<td>2.8×10^{-5}</td>
<td>This work</td>
</tr>
<tr>
<td>Suspended graphene</td>
<td>6×64</td>
<td>2.96×10^{-6}</td>
<td>Smith et al., Nano Lett. 2013</td>
</tr>
<tr>
<td>Graphene on fixed perforated layer on silicon substrate</td>
<td>110×220</td>
<td>0.88×10^{-6}</td>
<td>Hurst et al., Transducers, 2013</td>
</tr>
<tr>
<td>Carbon nanotubes</td>
<td>100×100</td>
<td>1.06×10^{-6}</td>
<td>Hierold et al., Sens. Actuator A, 2013</td>
</tr>
<tr>
<td>Silicon membrane</td>
<td>100×100</td>
<td>1.5×10^{-6}</td>
<td>Kalvesten et al., MEMS, 1998</td>
</tr>
</tbody>
</table>

**Conclusion**

In conclusion, we have demonstrated a graphene based small area MEMS pressure sensor formed by transferring large area CVD-grown graphene onto a suspended SiNx membrane perforated by an array of through-holes. The large voltage response of the sensor was majorly due to the large strain change of the graphene suspended over the through-holes under applied differential pressure across the membrane.
The measured sensitivity has demonstrated that the devised new pressure sensor structure excels in providing high sensitivity that outperforms many other existing graphene based counterpart sensors. Future work includes optimizing fabrication processes to reduce number of pinholes in graphene, improving yield of transferring graphene membrane to suspended nitride membrane, and designing a low-noise electronic readout circuit for the sensor.

**Methods**

**Device fabrication.** The device fabrication started with a 3-in double side polished silicon wafer (p-type). A 200 nm thick SiN$_x$ layer was formed on both sides of the wafer by plasmon enhanced chemical vapor deposition (Figure 6(a)). Etch windows were then created on the back side of the wafer with photolithography and reactive ion etching of SiN$_x$ (Figure 6(b)). Subsequently, an array of 2.5 µm diameter holes were patterned in the SiN$_x$ layer on the front side of the wafer with the same method as that used in the last step (Figure 6(c)). After that, an anisotropic etch of silicon substrate with tetramethylammonium hydroxide (20.0 wt %, 78°C, Sigma-Aldrich, St. Louis, MO) was performed to create a suspended nitride membrane (490×490 µm$^2$) (Figure 6(d)). The wafer was then diced into 6×6 mm$^2$ pieces for the following processes. Commercially available CVD-grown graphene film on a 25 µm thick nickel foil (1×1 cm$^2$, University Wafer, Boston, MA) was used as the sensitive material of the device. Only one side of the foil was coated with graphene. To transfer the graphene film to the suspended nitride membrane, we used the poly(methyl methacrylate) or PMMA based transfer method following the protocol given in Ref. [18]. In this step, the nickel foil with graphene was drop-coated with PMMA (molecular weight ∼996 000 by GPC, Sigma-Aldrich, dissolved in chlorobenzene with a concentration of 46 mg/mL) (Figure 6(e)). The foil was then cured at 180 °C for 1 min, followed by etching away the nickel substrate by FeCl$_3$ solution (0.1 g/ml, Sigma-Aldrich, St. Louis, MO) for 20 hr (Figure 6(f)). After that, the PMMA-graphene stack was picked up and washed with deionized water, and
then, was placed on the SiN<sub>x</sub> membrane treated with oxygen plasma (Figure 6(g)). Finally, the PMMA substrate of the graphene film was etched by PMMA remover (Nano remover PG, MicroChem, Westborough, MA) (Figure 6(i)). Next, the graphene resistor was patterned in an oxygen plasma etcher with the help of a shadow mask made of aluminum prefabricated by a high-precision milling machine (Supra CNC Mill, CNC Masters, Irwindale, CA) (Figure 6(j)). Then, another aluminum shadow mask was machined and placed on the device to make gold contacts by e-beam evaporation of a 200 nm gold layer (Figure 6(k)). In these shadow mask based patterning, careful alignment between the shadow mask and the device was needed. Finally, the device was realized (Figure 6(l)).

![Figure 6](image)

**Figure 6.** Schematic of the fabrication processes for the device.

**Measurement setup.** The backside of the device was adhered to the outlet of an acrylic glass based microfluidic channel with structural adhesives. Air pressure was applied from the inlet of the air
channel using a programmable syringe pump (KDS210P, KD Scientific, Holliston, MA). A commercial differential pressure sensor (MPX5500DP, Freescale Semiconductor, Austin, TX) was used to measure differential pressures applied across the sensing membrane. A feedback circuit was used to enhance stability of the pressure control system. The output voltage signal of the commercial sensor was recorded by a data acquisition device (DI-245, DATAQ Instruments, Akron, OH) and then was converted to a differential pressure. The graphene sensor was connected into a Wheatstone bridge circuit as shown in Figure 2(a). An input DC voltage of 20 mV was applied across the bridge circuit. The small voltage was applied to avoid excessive heating of graphene. The output voltage from the graphene sensor was recorded with a digital multimeter (34401A, Agilent Technologies, Santa Clara, CA).

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References


