Supplementary Discussion 1 – Calculation of the TMCPS device performance

The sensitivity of the graphene polymer device was calculated using the equations described in the main text. The values used for the calculation were taken from Table 1 in the main text and Supplementary Table 1. First, the capacitance of each cell in the TMCPS was calculated. This consists of three sections, namely the SU-8 supported section, the suspended section and a section in touch-mode. In calculating the capacitance of each of these sections the following assumptions were made:

- The supported sections and touching sections are flat and stress free.
- The membrane deflection behaves like an isotropic circular plate with linear elasticity.
- The suspended section of the membrane has a constant gradient, as shown in the dotted black line in Figure 2.b of the main text.
- The density of materials is constant for each of the constituent materials and does not change under pressure.
- The membrane thickness is constant across the entire membrane and does not change under pressure.
- Immersion of the TMCPS in water does not alter mechanical properties of membrane.
| Polymer | Mechanical properties | Response to environment |
|---------|-----------------------|-------------------------|
|         | $E$ (GPa) | Elongation at break (%) | Coeff. of thermal exp. (ppm) | Water absorption (% by wt) |
| SU-8    | $4.2^1$ | $6.5^2$ | $52^3$ | $0.65^*^3$ |
| Parylene-C | $2.5^4$ | $200^6$ | $38^5$ | $0.14^*^5$ |
| PMMA    | $2.3^6$ | $2.5^7$ | $93^7$ | $0.3-0.4^7$ |
| PU      | $0.006^8$ | $4^9$ | $57^9$ | $0.3-8^9$ |

**Supplementary Table 1 | Mechanical properties of polymers used in TMCPS device.**

The capacitance of each of these sections was then summed to give the capacitance of a single cell. The TMCPS consist of a total of 28x28 cells and therefore the total capacitance of the array is given by multiplying the total capacitance of a single cell. In practice, an additional capacitance contribution is measured from the electrical interconnects and charge build up in the sensor housing. We note that this parasitic capacitance is significantly higher than that of the device due to the setup of the experiment requiring along electrical extension from the measurement equipment to the water tank. A breakdown of the capacitance calculations for each of the constituent sections of a unit cell and the entire array, $C_{array}$ including the parasitic capacitance, $C_{par}$ are given in supplementary Table 2.

| Unit cell | Device |
|-----------|--------|
| $C_{sup}$ | $C_{array}$ |
| Value    | Value |
| pF       | pF    |
| 0.41      | 574   |
| 0.05      | 56    |

| $C_{t}$  | $C_{par}^*$ |
|----------|-------------|
| Value    | Value |
| pF       | nF     |
| 12.2     | 12.7    |
| 2.3      | 5       |

| $C_{sus}$ | $C_{0}$ |
|-----------|---------|
| Value    | Value |
| pF       | nF     |
| 7.19     | 14.39  |
| 1.3      | 61     |

**Supplementary Table 2 | Capacitance calculations of the TMCPS device.**

*Parasitic capacitance measured in underwater experiment
**Stress stiffening of the membrane**

Stress stiffening is a geometrical nonlinearity that needs to be considered for thin structures that, in addition to bending stiffness, also have axial stiffness. Even for an ideal-clamped plate with uniform thickness that experiences a large deflection due to a uniform load, the calculation of the deflection is complicated. The exact calculation of the deflection of the plate requires the knowledge of the stress-state of the plate, which in turn depends on the deflection. This leads to a nonhomogeneous system of equations (Karman equations) that can only be confronted by successive approximation techniques.\(^\text{10}\)

We estimated the stress in the membrane in touch mode by applying a Finite Element Analysis (FEA) using Comsol Multiphysics 5.3 software. First we setup a model of the three layer graphene-Parylene-C-PU composite using the material properties and device geometries as shown in Supplementary Table 1 and Table 1 in the main text. We then simulated the deformation of the composite membrane at the transition into touch-mode (when the membrane first makes contact with the cavity base) and in full touch-mode, when a uniform pressure is applied to its surface. Both of these simulations are shown in Supplementary Figure 1a. The latter state (touch-mode) is approximately the configuration of the graphene-polymer TMCPS in equilibrium.

Further we used this numerical model to simulate the stress in the membrane. The simulation showed a maximum tensile stress of 7.6 MPa at the center of the clamped edge of the membrane and an average stress of 1.2 MPa when the membrane is at the transition point, as shown in Supplementary Figure 1b.

The FEA software used was limited to solving the membrane stress only in a fully suspended state (without touch constraints) and therefore was unable to analyse the membrane in touch-mode. Instead, we estimate the stress in touch-mode by comparing to previous FEA simulations on thick plates in touch-mode.\(^\text{11}\) These simulations showed that during touch-mode, the stress near the touch-point of the membrane remains relatively constant compared to the stress before touch-mode. However, the stress at the clamped edge of the membrane increases significantly. For comparison, we have plotted the estimated stress-profile of the graphene-polymer TMCPS membrane in the solid line in Supplementary Figure 1b. Considering these FEA simulations, the estimated maximum and average stress in the equilibrium configuration of the TMCPS is 16 MPa and 2.9 MPa respectively.

The maximum in-plane stress of the membrane was then simulated as a function of pressure up to the touch-point using out FEA model and further extrapolated using the previous study on TMCPSs. A plot of this simulation is shown in Supplementary Figure 1c. This model gives an approximate value of the critical slippage pressure of \(P_{\text{slip}} = 125 \text{ kPa}\), at which slippage is expected at the graphene-polymer interface (strain of 1.4%). Further we note that the estimated maximum stress is based on a model whereby the edges of the membrane are fully-clamped in the plane of the membrane. In reality, the edge of the SU-8 spacer has a radius of curvature of approximately 20 µm, suggesting that the stress at the edge of the membrane is distributed over a larger area and therefore lies a lot closer to the average value of stress than the maximum as previously predicted.

The effect of stress stiffening has an overall effect of decreasing the deflection sensitivity of the membrane. For the estimated stress of the graphene-polymer membrane in touch-
mode this effect reduces the sensitivity of the device by approximately 7%. This stress stiffening effect has been considered in the sensitivity calculation of the device.

**Supplementary Discussion 2 – Fabrication protocol of TMCPS device**

Sensor fabrication begins with a graphene flattening process (Figure 2, step 1). A copper foil of 15 mm x 15 mm size with CVD-graphene on its top surface is coated with a thin Parylene-C layer (Figure 2, step 1a). The Parylene-C (Di(2-chlor-p-xlylen)) is deposited by chemical vapor deposition using a SCS Labcoater 2010 system with an Inficon XTC2 deposition controller. For the entire deposition the pyrolyse furnace is set to 690°C. The deposition chamber is pumped to 15mTorr before the vaporiser is switched on and heated up to 175°C. The deposition is finished when the temperature (175°C) is reached and the deposition rate is stable. The samples are kept in the chamber during the chamber cooling step. The thickness of the Parylene-C is given by the initial dimer weight portion. This produced conformal coatings of $23 \pm 5 \text{ nm}$, $101 \pm 8 \text{ nm}$, $218 \pm 17 \text{ nm}$ and $510 \pm 35 \text{ nm}$ of Parylene-C on
the CVD graphene surface. The foil is then floated in a 2.5 wt/vol % aqueous ammonium persulfate solution for 4 hours in order to etch away the copper (Figure 2, step 1b). The remaining graphene-Parylene-C membrane is then transferred into a deionised (DI) water bath by fishing it with a microscope slide. This process is repeated for two further DI water baths, in 15 minute intervals to allow contaminants to be cleaned from the graphene surface (Figure 2, step 1c). After the third DI water bath the film is transferred onto a plasma cleaned Si/SiO$_2$ substrate using the same fishing method (Figure 2, step 1b). As the water dries from this substrate the graphene-Parylene-C stack conforms to the surface of the SiO$_2$.

The second step in the sensor fabrication is to deposit an additional flexible polyurethane (PU) layer and release a homogenous graphene-polymer heterostructure from the substrate (Figure 2, step 2). A 10 µm thick PU layer (Hyperlast LU 1009, Dow Chemicals) is bar coated onto the graphene-Parylene-C membrane and left to cure for 24 hours (Figure 2, step 2a). The substrate is then submerged in an aqueous potassium hydroxide solution (30 wt%) for up to 5 hours until the CVD-graphene-Parylene-C-PU membrane lifts off the substrate and floats on the surface of the etchant solution (Figure 2, step 2b). Similarly to step 1, the membrane is cleaned in 3 subsequent DI water baths before it is removed from the final bath with tweezers (Figure 2, step 2c).

The next step is to form the target substrate comprising cavities and electrodes (Figure 2, step 3). On a separate Si/SiO$_2$ substrate, a 10 µm thick layer of SU-8 2025 (MicroChem) positive photoresist is deposited by bar coating. The SU-8 layer is exposed and developed according to the step described in Table 3 using a MJB4 mask aligner and a custom Cr/Glass photomask (UVLithoSys inc.). A photograph of the sensor chip at this stage of the fabrication process is also shown in Supplementary Figure 3. A small bead of silver epoxy is then deposited onto the corner of the SU-8 spacer pattern and onto the side of the chip. The silver epoxy has a curing time of up to 16 hours, in which time electrical connection leads are inserted into the epoxy and the graphene-polymer laminate is deposited onto the chip.

Supplementary Figure 2 | Fabrication steps and schematics of a TMCPS.
The final step of the fabrication process is the transfer of the graphene-polymer membrane on to the silicon chip (Figure 2, step 4). This process is carried out using a custom alignment system win which the graphene-polymer membrane is freely suspended above the target substrate and then lowered down onto it. Once the membrane and chip have made contact, a subtle force (~5 kPa) and an elevated temperature (65°C) are applied to the device in order to giver good adhesion between the membrane and the chip. A photograph of the final device is shown in Supplementary Figure 2.b.

**Supplementary Table 3 | Processing steps of SU-8 positive photoresist.**

| Step   | Description (part 1)          | (part 2)         |
|--------|--------------------------------|------------------|
| 1.     | Post exposure                  | Bake at 95°C for 5 min. |
|        | Bake at 65°C for 1 min.        |                  |
| 2.     | Exposure                       | Expose with substrate with UV lamp for 160 secs. |
| 3.     | Post exposure                  | Bake at 65°C for 1 min. |
|        | Bake at 95°C for 5 min.        |                  |
| 4.     | Development                    | Agitate in EC solvent for 5 mins. |
|        | Rinse in IPA                   |                  |

**Supplementary Figure 3 | Photographs of sensor chip (a) during fabrication (step 3.b. in reference to Supplementary Figure 2) and (b) after fabrication (on a glass backing and steel backplate)
Supplementary Discussion 3 – Raman spectroscopy of TMCPs device

The Raman spectra of bare graphene and 12 µm thick CVD-graphene/Parylene-C/PU supported on Si/SiO$_2$ substrates were measured at an excitation energy of 2.4eV (514 nm) using an inVia™ Renishaw confocal microscope. The spectrum of the graphene-polymer membrane is representative of single-layer CVD graphene, with a distinctive G and 2D peak at around 1580 and 2680 cm$^{-1}$ respectively. Compared to bare CVD-graphene, we observe a drop in the intensity ratio between the G and the 2D peak as well as emerging of the D peak (1340 cm$^{-1}$)$^{12}$. This is suggestive of some defects in the graphene layer, however, previous reports on bare single-layer CVD graphene have shown a similar degree of defects whilst demonstrating the same superior mechanical characteristics of pristine graphene$^{13}$. Thus, we assume that the deposition of Parylene-C and PU have negligible effect on the structural integrity of the graphene layer. In addition we did not observe any significant shift in the G or 2D peak as a function of polymer thickness, which is consistent with previous measurements$^{14}$.

Supplementary Discussion 4 – Pressure sensor calibration

In order to demonstrate the effect of the water environment of the capacitance of the sensor device we measured we measured the capacitance of the TMCPs device when positioned just below the surface of the water such that minimal pressure is acting on the membranes. The capacitance of the device over the course of an hour is shown in Supplementary Figure 5. This shows a gradual increase in capacitance during the first 30 minutes of submersion. This increase in capacitance is attributed to partial permeation of water molecules into the matrix of the PU. After approximately 30 minutes the capacitance plateaus and remains stable with an accuracy of 0.1% as shown in Supplementary Figure 5b. Thus, all pressure cycling measurements taken throughout this study were gathered after an a 30 minute period in water immersion had elapsed.
Supplementary Discussion 5 – Dynamic response of TMCPS device

As a proof of concept for underwater acoustic sensing, a glass beaker filled with silicon oil and a TMCPS device mounted inside, was tapped with rubber tool to induce pressure waves in the oil container. The pressure of the tap on the outer surface of the beaker was estimated to be between 100 kPa and 200 kPa. A dummy device without cavities, showing a negligible response to the tap, confirmed that the pressure pulse does not influence the measurement setup other than the TMCPS membranes. Supplementary Figure 6a and 6b show the response of cavity containing TMCPS with cavities in the time and frequency domain respectively. The time resolution of the impedance analyser used in this experiment was 1 ms therefore giving a frequency range limited below 500 Hz.

Supplementary Figure 5 | Calibration of pressure sensing system. (a) Variation of capacitance as a function of time after immersion of the pressure sensor into the water tank. (b) The change in capacitance of the TMCPS device after 30 minutes is rescaled to highlight the accuracy of the capacitance measurement.

Supplementary Figure 6 | Dynamic response of TMCPS device. a) Response to tapped container pulse of graphene-polymer CMUT in oil immersion. b) Fourier transform of the receive signal in Figure 5a converted to dB relative to the peak signal 478 Hz.
Supplementary Discussion 6 – Demonstration of force sensor in air

In order to demonstrate the application of the TMCPS device as a force sensor in air, we conducted a simple experiment where the sensor was pressed with a finger. In this experiment the sensor was packaged in polyimide tape to ensure the force of the finger does not influence any of the electrical connections to the wire. Supplementary Figure 7 shows the response a simple touch of the top surface of the sensor that is estimated to exert a force of 1N. A demonstration of this simple experiment is also given in a Supplementary Video.

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