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SUPPLEMENTARY INFORMATION
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Ultra-strong Adhesion of Graphene Membranes

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Supplementary Information:

Counting Number of Graphene Layers

In order to count the number of graphene layers used in this study we employed a combination of optical contrast, Raman spectroscopy, AFM measurements, and the elastic constant measurements. Raman spectroscopy has been demonstrated to be a powerful tool for identifying single layer graphene sheets [1]. Recently Raman has also been shown to be able to identify the number of layers of few layer graphene, a technique we use here[2]. Figure S1 (a) and (b) show the graphene flakes from this study and the spots where Raman spectrum was taken for each device, black is 1 layer, red is 2 layers, green is 3 layers, blue is 4 layers and cyan is 5 layers. Figure S1 (c) and (d) show the Raman spectrum taken from the spots of corresponding color in (a) and (b) respectively. To verify the number of layers we found the ratio of the integrated intensity of the first order optical phonon peak and the graphene G peak. The ratios are shown in figure S1 (e) and (f). Comparing these values with the Fresnel equation we can determine the number of layers for each region. In order to verify this technique we used optical contrast, AFM measurements, as well as the elastic constants of the membranes [3]. The optical contrast and AFM measurements showed close agreement to the Raman spectroscopy technique validating its utility.

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Adhesion Energy and Elastic Constants Measurements

The adhesion energy measurements were carried out according to the main text of this article. Figures S2, S3, and S4 show (a) δ vs. p_0, (b) a vs. p_0, and (c) p_{int} vs. p_0, for all the membranes studied. The layer numbers are as follows: (a) 1 layer membranes from Fig. 1 (lower). (b) 2 layer membranes from Fig. 1 (upper). (c) 3 layer membranes from Fig. 1 (upper). 4 layer membranes from Fig. 1 (upper). (d) 5 layer membranes from Fig. 1 (upper). and (e) 3 layer membranes from Fig. 1 (lower).

Repeatability of Elastic Constant Measurements

To verify the repeatability of the measurement of the elastic constants at $\Delta p < 0.5$ MPa we first pressurized the graphene flake in Fig. 1a(upper) up to $\Delta p =$ 0.45 MPa and then let pressure decrease back to $\Delta p =$ 0 MPa. We then repeated the measurements and increased Δp until there was significant peeling from the substrate in order to test the adhesion strength. Figure S5 shows the results from this test for (a) 2 layers, (b) 3 layers, (c) 4 layers, and (d) 5 layers of graphene. From this we conclude that pressurizing the membranes does not cause sliding or change the membrane properties when $\Delta p < 0.5$ MPa and therefore the membrane can be considered to be well clamped to the substrate in this pressure range.

Adhesion from Trapped Charges in SiO_2

We use the method of image charges to estimate the influence of trapped charges in the SiO_2 on the adhesion of graphene to the substrate. The work needed to move a charge from a distance d from the conducting plane out to infinity is:

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$$W = \frac{1}{4\pi\varepsilon_o}\frac{q^2}{4d} \quad \text{(S1)}$$

where q is the fundamental charge, d is the distance the charge is away from the conducting plane and ε_o is the permittivity of free space[4]. In order to determine an adhesion energy we also need to know the area density of charges, ρ, and the equation becomes:

$$\Gamma = \frac{1}{4\pi\varepsilon_o}\frac{q^2}{4d}\,\rho \quad \text{(S2)}$$

If we assume all the charges are on the surface of the SiO_2 and that the equilibrium spacing between the graphene and SiO_2 is equal to that of the equilibrium spacing of graphite d = 0.34 nm. The charge density needed to produce our measured adhesion energy of 0.31 J/m^2 is ~9x10^{17} m^{-2}. The charge density of SiO_2 is reported to be 2.3x10^{15} m^{-2} [5]. Seeing that the reported value of the charge density in SiO_2 is almost three orders of magnitude lower, we can conclude that trapped charges do not have a significant contribution to the adhesion energy value we measure. Other studies have used potassium ions to increase the charge density present in the oxide [6]. The concentration of potassium ions was as high as ~5 x 10^{16} m^2. This upper limit of the extrinsic doping concentration results in a charge density that is one order of magnitude less than that needed to have adhesion energies on the order of what we measured. These results show that the effect of charge impurities in the SiO_2 below the graphene will not significantly influence our measure of adhesion energy.

RMS Roughness and Conformation

Roughness measurements were taken using a Veeco Dimension 3100 operating under non-contact mode under ambient conditions. The bare SiO_2 substrate is denoted as 0 layers in Fig. S6 and a ~5nm thick flake as measured by the AFM was estimated to be

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approximately 15 layers thick. For the roughness measurements of the substrate and each layer thickness multiple images were taken at various locations of each region, the images were taken from the chip in Fig. 1a (lower) and the RMS roughness was analysed using Wsxm software for each image [7]. The 1-3 layers were taken from the flake in Fig. 1a while the substrate measurements were taken from areas around the flake and the ~15 layer measurement was taken from a thick flake near the flake seen in Fig. 1a(lower). For the substrate and each different layer thickness, 7 images were used for the substrate, 4 images were used for 1 layer, 5 images for 2 layer, 3 images for 3 layers, and 2 images for the ~15 layer sample. Figure S6 shows the average roughness for the substrate, 0 layers, 1 layer, 2 layers 3 layers and ~15 layers as well as the standard deviation of the measurements shown by the error bars. These measurements suggest that graphene conforms more intimately to the substrate and as the number of layers is decreased

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Supplementary Information Figures

Figure S1. **Counting the Number of Layers**

(a) and (b) Optical images showing the graphene flakes used in this study. The colored circles denote the location at which Raman spectroscopy was taken (denoted as follows: black 1 layer, red 2 layers, green 3 layers, blue 4 layers, and cyan 5 layers)

(c) and (d) Raman spectrum from the graphene flakes in (a) and (b). The color of each curve corresponds to the spot on the optical image.

(e) and (d) Ratio of the integrated intensity of the first order silicon peak I(Si) and graphene G peak, I(G) (i.e. I(G)/I(Si)).

Figure S2. **Measured Deflection vs. Input Pressure**

(a) – (f) δ, vs p_o, for 1-5 layer devices. 1 layer devices (a) are from graphene flake in Fig. 1a(lower) and the 2-5, (b)-(e) respectively, are from the flake in Fig. 1a. (f) The data in f was determined to be 3 layers thick and taken from the lower graphene flake in Fig 1a.

Figure S3. **Blister Radius vs. Input Pressure**

(a) – (f) a vs. p_o for 1-5 layer devices in Fig. S2.

Figure S4. **Internal Pressure vs. Input Pressure**

(a) – (f) p_{int} vs p_o for 1-5 layer devices in Fig. S2.

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Figure S5. **Repeatability of Measurements at Low Pressure Differences**

(a) - (d) $K(\delta^3/a^4)$ vs Δp for 2-5 layer devices. The black points are from the first pressure cycling of the upper device in Fig. 1(a). After the highest pressure was measured the pressure was allowed to decrease back to atmospheric pressure and the measurements were repeated and carried higher pressures. This shows that up to $\Delta p \approx 0.5$ MPa there is no altering of the membrane properties between measurements.

Figure S6. **Measured Roughness of the Substrate**

RMS roughness measurements taken by non-contact AFM of the substrate (0 layers), 1, 2, and 3 layers as well a thick graphene sample that was ~5 nm (~15 layers) thick as determined by the AFM. Error bars are ±1 standard deviation.

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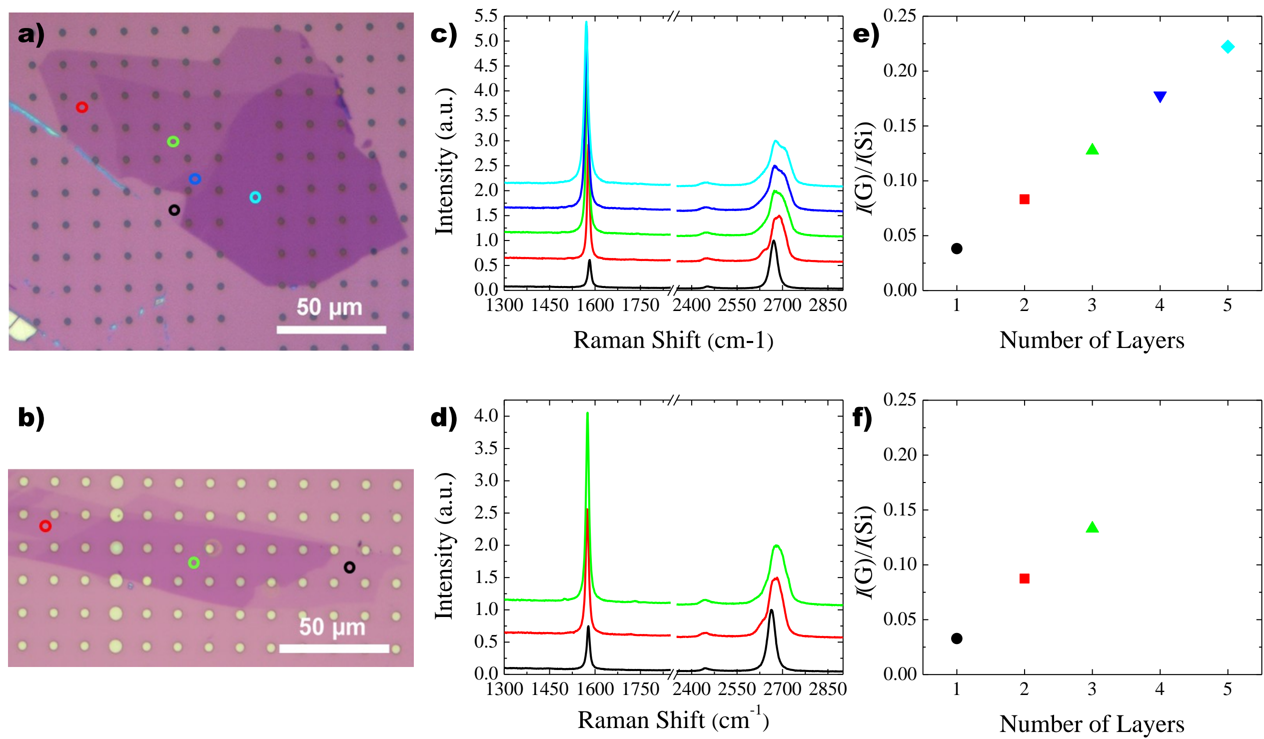

Figure S1

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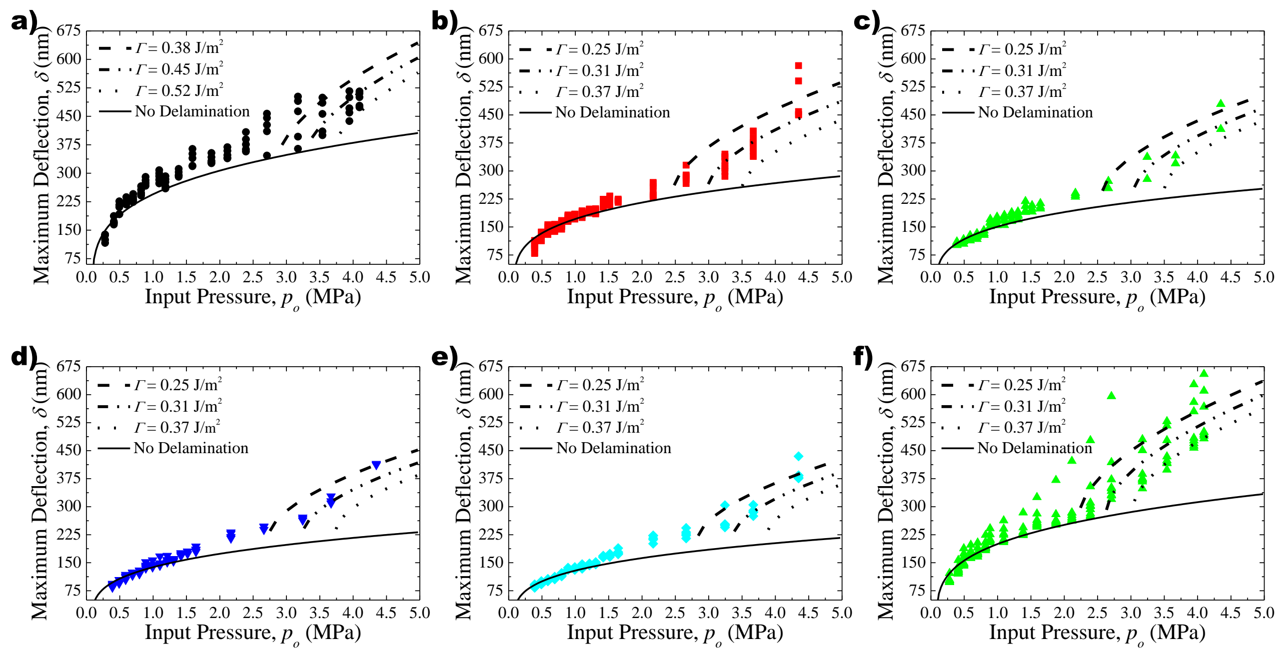

Figure S2

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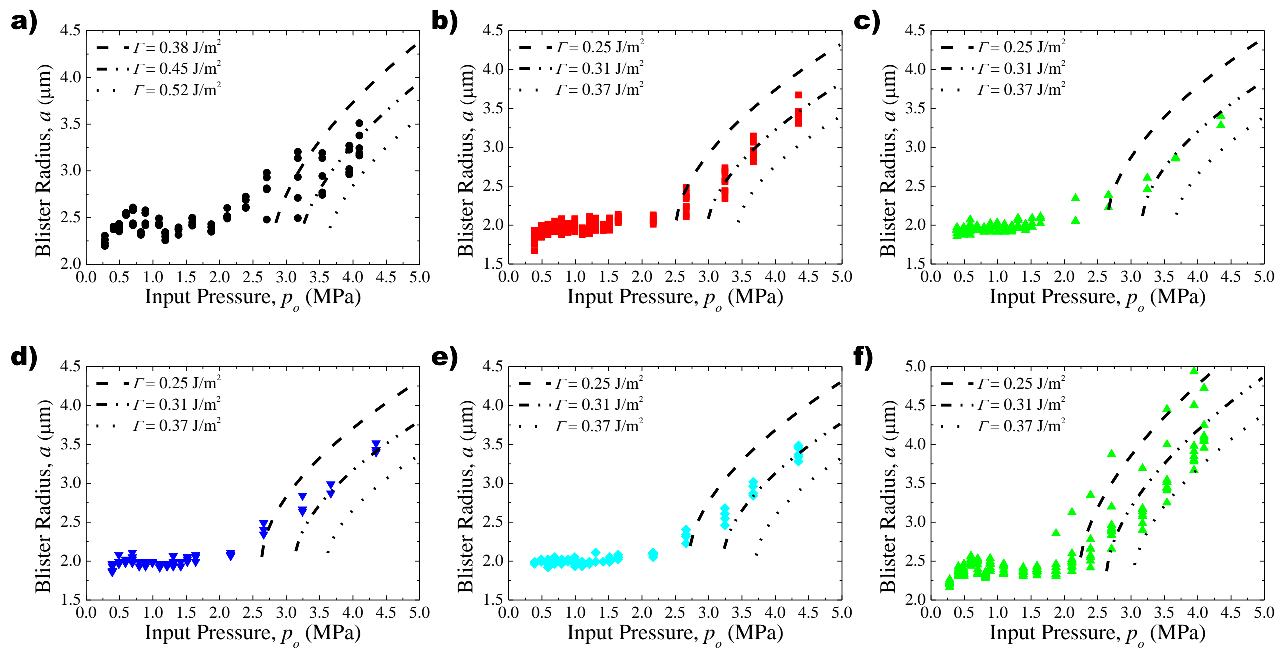

Figure S3

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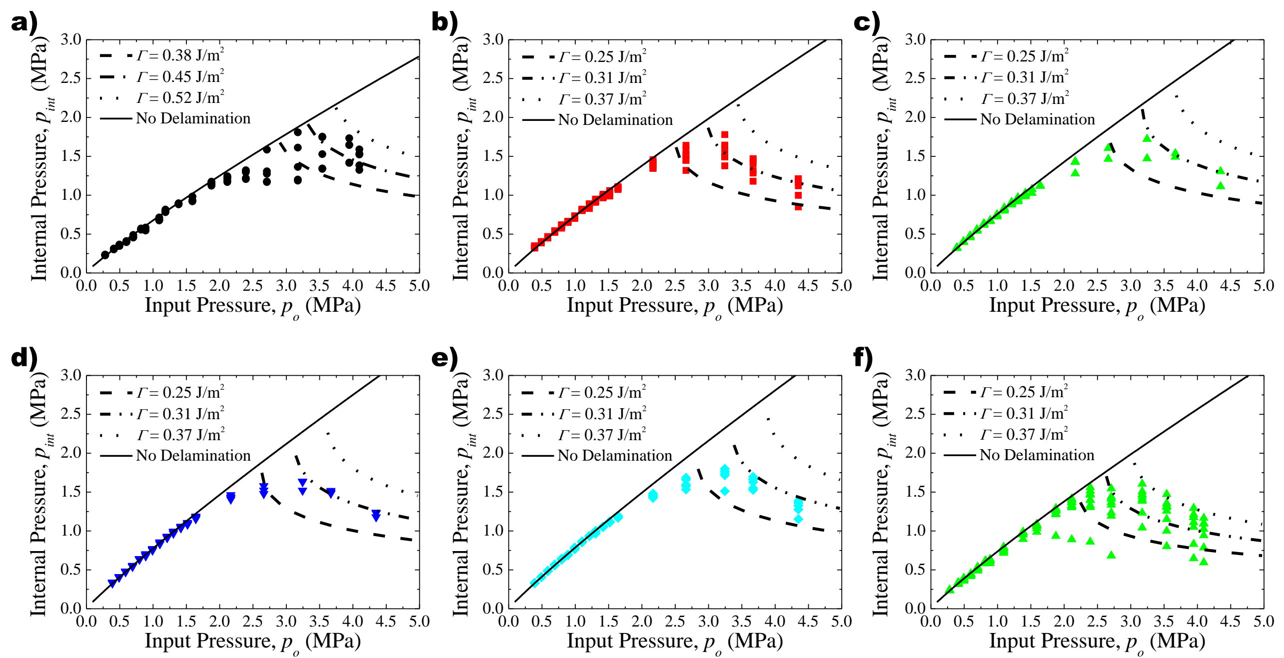

Figure S4

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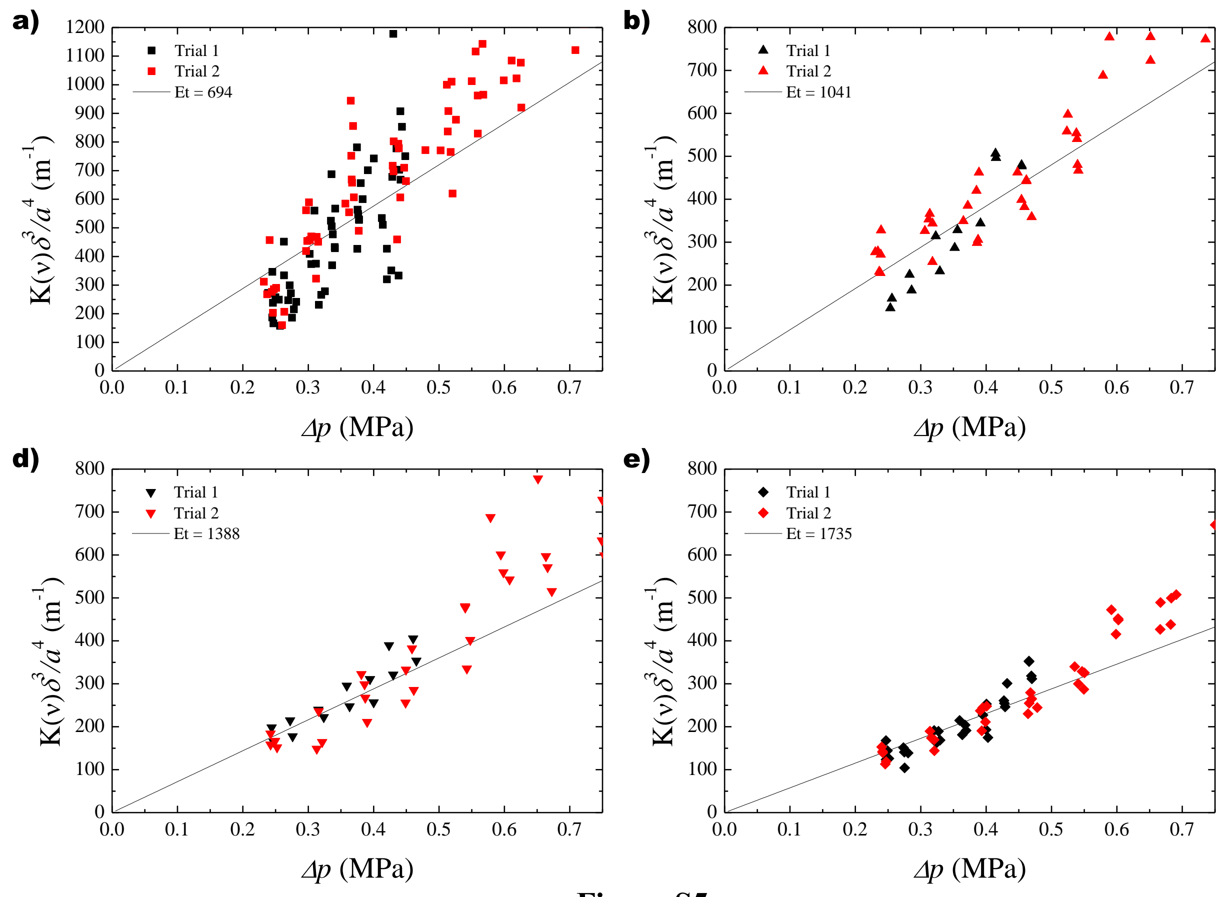

Figure S5

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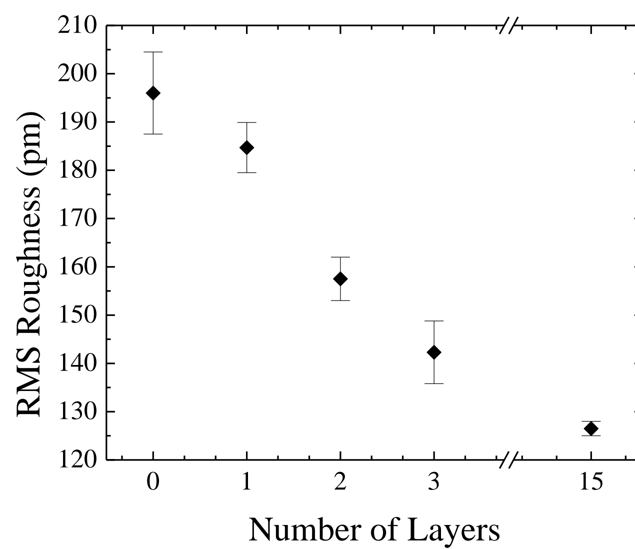

Figure S6

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