Supporting Information

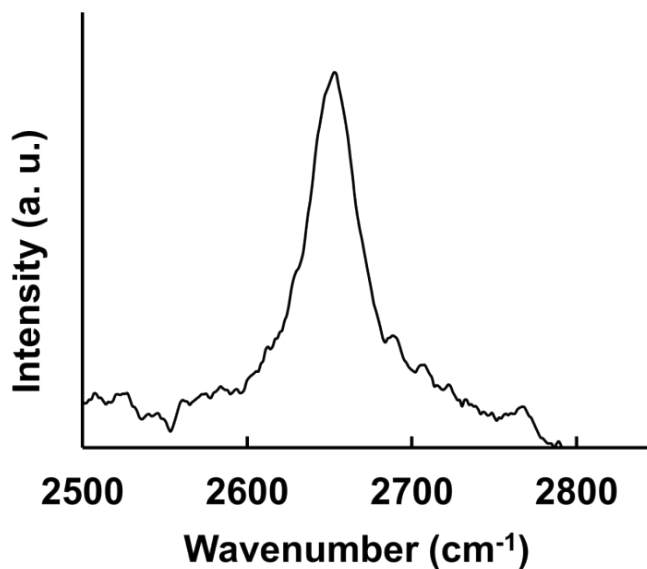## 1. Raman spectrum of monolayer graphene

Figure S1: Raman spectrum of suspended graphene before TiO_2 deposition

Raman spectroscopy was conducted with a Renishaw inVia Confocal Raman Spectrometer using a 514 nm laser. A single 2D peak at ~2700 cm^{-1} is a signature of monolayer graphene. Such a sharp peak is not present for multilayered graphene that exhibits a broader peak with a shoulder[1].

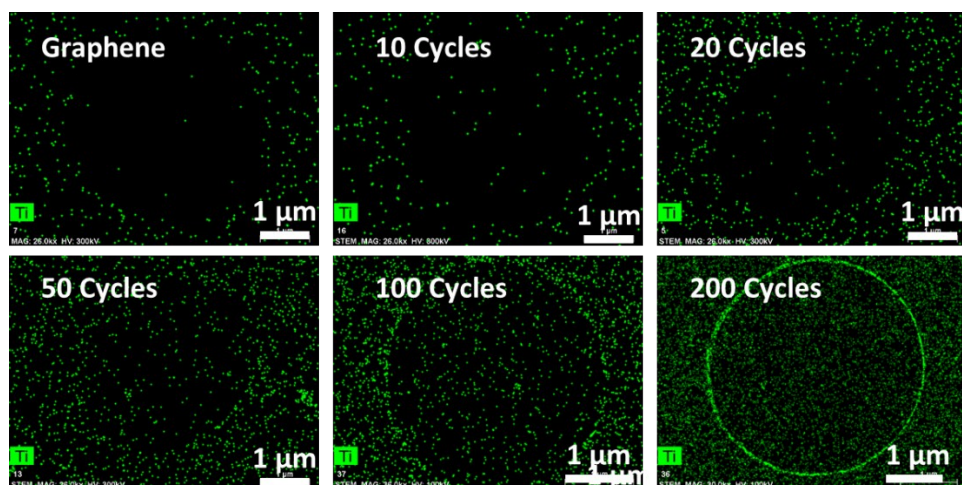## 2. TEM Electron Energy-Dispersive X-ray (EDS) Elemental Mapping

Figure S2 Titanium EDS elemental mapping of different samples.

Scanning transmission electron microscopy (STEM) and energy-dispersive X-ray spectroscopy (EDS) were performed using a Hitachi HF-3300 TEM/STEM at 300 kV and 100 kV. EDS elemental mapping was used to monitor the coverage of titanium on graphene. With an increasing number of ALD cycles, more titanium was deposited. There is no significant titanium coverage on graphene and 10 ALD cycles prepared samples, and any apparent Ti signal is within the noise limit of the detection. For 20 & 50 ALD cycles-prepared hetero-films, a partial titanium coverage was observed. When graphene was deposited for 100 & 200 ALD cycles, titanium completely covered the graphene surface.

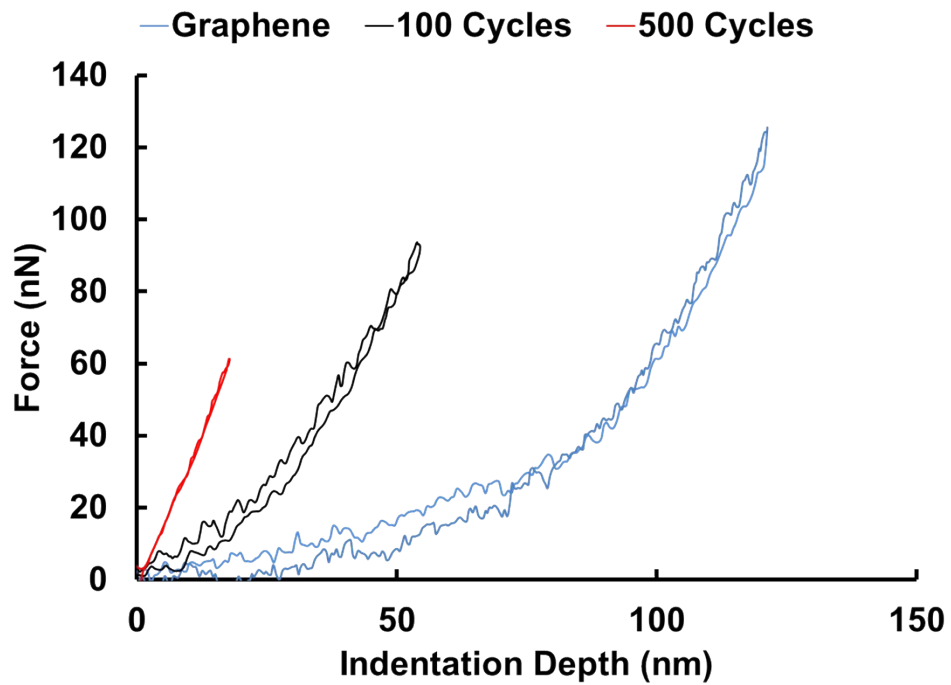# 3. Representative curves: loading/unloading force versus indentation depth

Figure S3: Representative loading/unloading curves of graphene, 100 and 500 ALD cycles prepared TiO_2/graphene hetero-films. No significant hysteresis was observed between the loading/unloading curves.

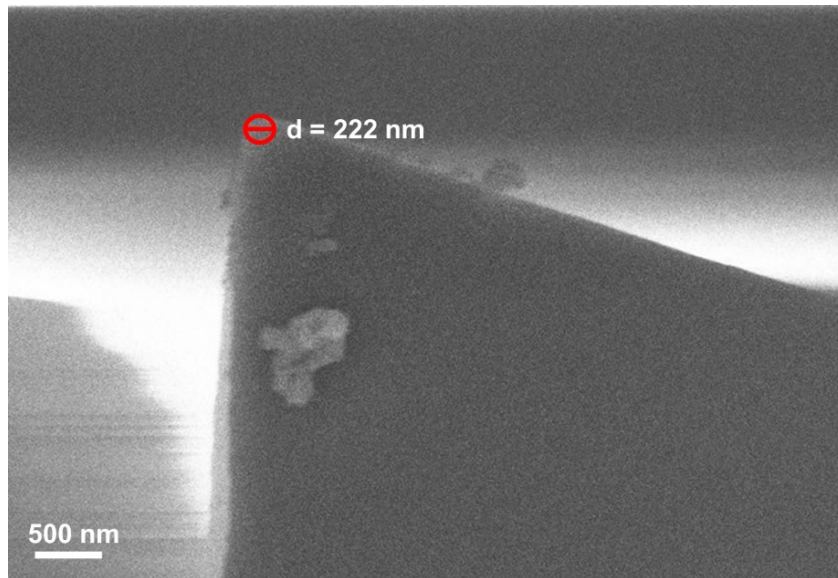## 4. Diamond AFM tip diameter measurement

Figure S4: SEM image showing the measurement of the diamond AFM tip diameter
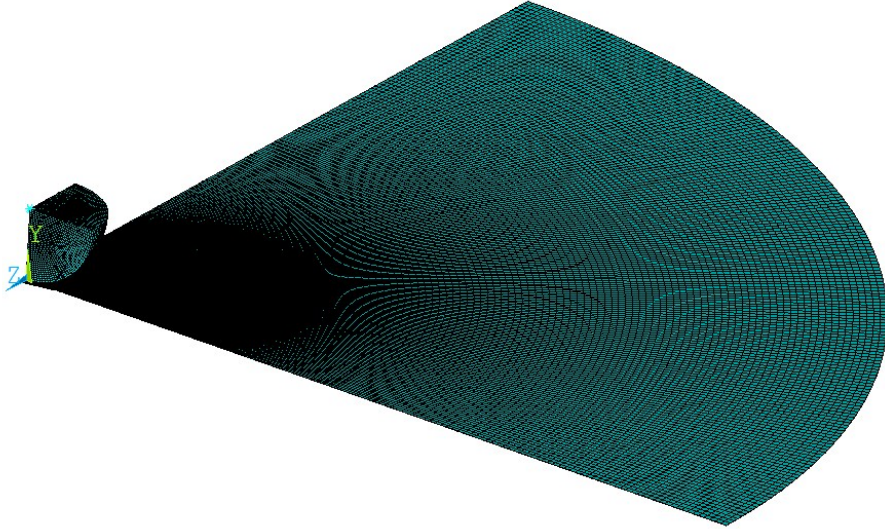
5. Finite element modelling (FEM)

Figure S5: Mesh of a quarter of a 2.5 µm diameter TiO_2/graphene hetero-structure in finite element analysis

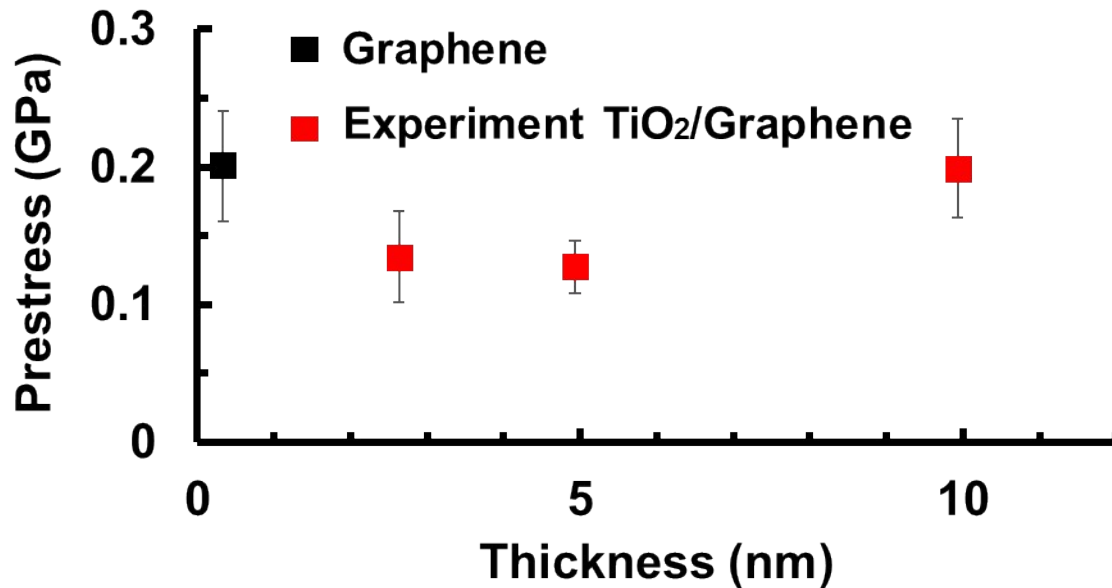## 6. Prestress versus TiO$_2$/graphene thickness

Figure S6: Prestress in monolayer graphene and TiO$_2$/graphene hetero-films as a function of thickness

In Figure S6, prestresses are plotted as a function of thickness. It can be seen that the pretension is independent of their thicknesses. The maximum fluctuation in pretension is observed in the thickest structure, i.e. the 24.9 nm sample (500 ALD cycles). The source of prestress in these thin structures is unclear; however, likely, the stress is induced during the processing of the graphene monolayer and the atomic layer deposition of TiO$_2$.

References:

[1]   A. C. Ferrari, J. C. Meyer, V. Scardaci, C. Casiraghi, M. Lazzeri, F. Mauri, S. Piscanec, D. Jiang, K. S. Novoselov, S. Roth, A. K. Geim, Physical Review Letters 2006, 97.