Supplementary Information

Probing the interaction between 2D materials and oligoglycine tectomers

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Figure S1. Tectomer deposited on MoS$_2$ and graphene: (a) High-resolution AFM morphology of tectomer-covered MoS$_2$; inset illustrates the thickness of the tectomer 5.6 ± 0.5 nm sheet. (b) The high-resolution AFM image of a flat graphene surface illustrates the thickness of the tectomer 5.6 ± 0.5 nm sheet.
Figure S2. Thickness of MoS$_2$ and graphene: (a) AFM topography of the individual monolayer MoS$_2$ island, (b) multilayer MoS$_2$ island. (c) Topography of CVD graphene transferred over silica, dashed line is drawn at random torn region occurred during ultrasonic bath cleaning. (d-e) The corresponding line profile of the dashed region in panel (a)-(c) respectively. It is worthy to note that thickness distribution from AFM is influenced from substrate roughness, conformation of graphene, trapped air and water molecules during transfer and AFM probe deflection instability moving over hetero-surfaces (silica-graphene). (g, h) $E_{12g}$ and $A_{1g}$ Peak shift distribution over an ML MoS$_2$. (i) Difference in the frequency shift ($A_{1g}$-$E_{12g}$) to depict the thickness of MoS$_2$ sheet.
Figure S3. Histogram of peak positions and strain and doping: (a) Data distribution between 1L MoS$_2$ and tectomer/1L MoS$_2$ in terms of E$^{1g}$ peak position, (b) A$_{1g}$ peak position, (c) strain and (d) doping. (e) Distribution of data between 1L Gr and tectomer/1L Gr in terms of G peak position, (f) 2D peak position, (g) strain and (h) doping.
Figure S4. High resolution Mo3d, S2s and S2p XPS spectra of (a) pristine MoS$_2$ and (b) tectomers/MoS$_2$
Figure S5. TEM (a, b), SEM (c) and EDX (d, e, f) of tectomer (T)/MoS$_2$ hybrids. The higher contrast of MoS$_2$ nanosheets in TEM images (a, b) enables distinguishing both components in TEM micrographs. It can be noticed that there are no MoS$_2$ nanosheets on the TEM grids, but only on tectomer platelets, therefore suggesting strong interaction between both materials.
Figure S6. TEM micrographs of tectomer platelets (a, b). TEM (c, d) and SEM (e, f) characterization of graphene covered tectomer platelets.
Figure S7. Electrical characterisation of mechanically exfoliated graphene (few layers) and MoS$_2$ (few layers) on interdigitated electrodes (IDEs): (a) The trend of resistance of ML graphene on IDEs (Interdigitated Electrodes/ Microelectrodes) as a function of time. The blue arrow indicates the time interval when tectomer molecules were deposited over ML graphene, and the brown dash line indicates the time interval where tectomer analyte is dried (approximately 2 hours). (b) The corresponding I-V curve of the graphene IDEs. Conductivity decreases after exposure to tectomer. Given that graphene is usually p-type on silica, it suggests n-doping from tectomer to ML graphene. (c) The resistance of ML MoS$_2$ on IDEs as a function of time. The blue arrow indicates the adsorption of tectomer and the brown dash line indicates the time interval where tectomer analyte is dried. (d) The corresponding I-V curve of the MoS$_2$ IDEs. Conductivity increases after exposure to tectomer. Given that MoS$_2$ is usually n-type, it suggests n-doping from tectomer to ML MoS$_2$. 
**Figure S8.** (a) Schematic diagram showing the interaction between Pt/Ir coated tip apex with amino groups of tectomer for different time intervals of 0.2 to 50 seconds at constant applied load. (b) Schematic view of a force-distance spectroscopy showing approach (green color) and withdraw (blue color) of cantilever. There is a pull-out moment (marked by black arrow) that shows work done by the tip to counter the adhesion force from the tectomer surface. (c) The increase of adhesion force (nN) at different duration of contact between tip apex and tectomer.

The surface chemistry of the 2D material can be described by the adhesion force (pull-out force) between the surface and the platinum/iridium (Pt/Ir) AFM tip. It is work done by the cantilever to move away from the surface which can be illustrated by the AFM Force-Distance curve in Fig. S6a, b, where the adhesion force represents the attraction force to obstruct the withdrawal of the tip from the material as labelled by the vertical arrow in the graph. As the adhesion force is time-dependent, we applied a constant load (nN) to tectomer with different durations (seconds) (Fig. S6b) and observe a logarithmic relation between the adhesion and the contact time (Fig. S6c), which is supported by a recent work of Lai et al. In their work, the adhesion force $F$ can be expressed as

$$F = a + b \cdot \log \left( \frac{t_{\text{contact}}}{t_0} \right)$$

where $t_{\text{contact}}$ is the contact time, $t_0 = 30$ s is the dwell time during contact, while $a$ and $b$ depend on environmental governing factors such as vapour pressure and humidity. This trend indicates the adhesion force increases with duration, but saturation occurs when the contact time falls into the regime beyond 30 s. The interaction between Pt/Ir tip and tectomer is pivotal to prepare the functionalized tip and it should be higher than tectomer-2D materials for the durability of the coatings during AFM operations.

Figure S9. Studies on adhesion affinity for different number of tectomer layers: (a) AFM topography and (b) its corresponding adhesion map of tectomer on graphene with different number of layers. (c) The height and adhesion profile of the dashed line shows no significant difference in adhesion force for different tectomer thickness.
Figure S10. Detection of the coated tectomer mass over cantilever: (a) SEM micrograph of a cantilever without tectomer attached. (b) SEM micrograph of the same cantilever partially coated with tectomer (black coloured region). (c) The change in resonance frequency of the same cantilever for pre and post-deposition of tectomer reveals a mass increment.

The Resonance frequency changes due to increase in mass of the cantilever by the given relation:

$$4(RF)^2 = K/M_{\text{eff}}$$

where, RF is the resonance frequency and $M_{\text{eff}}$ is the effective mass.

Assuming the same stiffness of the cantilever, the addition of mass ($M$) over cantilever is evaluated as follows:

$M$ (mass of the cantilever beam) = $\rho \times \text{vol} = 3100 \, \text{Kg/m}^3 \times 21265.2 \times 10^{-18} \, \text{m}^3 = 6.59 \times 10^{-8} \, \text{g}$

$M_{\text{eff1}}$ (effective mass during cantilever vibration) = $133/40 \times (M) = 2.19 \times 10^{-7} \, \text{g}$

Also, $(RF1/ RF2)^2 (M_{\text{eff1}}) = M_{\text{eff2}}$ (mass of the coated cantilever)

$M_{\text{eff2}} = 2.49 \times 10^{-7} \, \text{g}$

Mass of coating over entire cantilever = $\Delta M = M_{\text{eff2}} - M_{\text{eff1}} = 2.9 \times 10^{-8} \, \text{g} = 29 \, \text{ng}$
Figure S11. QNM indentation maps: The indentation of (a) graphene and (b) MoS$_2$ is measured to be lower than the silica substrate, indicating the adhesion contrast is solely due to the chemical interaction instead of mechanical indentation.
Figure S12. (a) Adhesion force map of tectomer-coated tip on silica which is doped from -2V to 2V. The doping is carried out by applying voltage bias on the AFM stage which is connected to the silica by carbon tape. (b) Line profile of the corresponding adhesion force shows that the adhesion between tectomer and silica increases with increasing induced charges, suggesting tectomer is more susceptible to donate electrons.