Supporting Figures:



Supplementary Figure S1 - Hopp and Woods hydropathy (ExPaSy tools) plot of Mfp-5 using hydropathy values from (Nozaki and Tanford) for standard amino acids and Dopa. Phosphoserine hydropathy was assumed to resemble values for aspartate.



Supplementary Figure S2- Effect of salt concentration on the Mfp-5/Mfp-5 self-interaction (i.e. cohesion) after a 14 min (840 s) wait time. Deposition of Mfp-5 of symmetric mica sheets at pH 2.6 and I=100mM. Circled portions indicate peak adhesive strengths F_{ad} of the run.



Supplementary Figure S3- Mefp-5 adhesion to mica showing adhesion abolition upon raising the pH to 7.5. I=100mM and I=584 for pH 2.6 and pH 7.5, respectively.



Supplementary Figure S4- Mefp-5 adhesion to mica as plotted on a Log/Log plot with the total contact time against the adhesive force. Linear regression slope= 0.103, R²=0.82.

Supplementary data - Calculation of the refractive index and volume fraction of compressed protein:

Calculation of the volume fraction of protein in the compressed layer between two mica surfaces in SFA is possible if the refractive index μ can be determined:

$$\mu = \mu_{mica} \sqrt{\frac{(n-1)F_{n-1}}{nF_n} * \frac{\lambda_{n-1}^D - \lambda_{n-1}^0}{\lambda_n^D - \lambda_n^0}}$$
 (for odd n) (equation A4 from reference 18)

where μ_{mica} is the refractive index, λ is the wavelength, n is fringe order and F_n is a correction factor approximated by $F_n = 1.024 + 1/n$. Calculations of 16 runs from three different set-ups gave a refractive index of 1.538 ± 0.012 . Using the knowledge that refractive index of water (μ_w) and pure protein (μ_p) have assumed values of 1.333 and 1.55 respectively, the simple formula

volume fraction
$$= \frac{\mu - \mu_w}{\mu_p - \mu_w}$$

This gives the 95% Mefp-5 \pm 5% by volume in the compressed layer.

Supplementary data - Calculations for protein deposition and H-bond strength:

We can use the average $\frac{1}{4}cm^2$ piece of mica with a protein deposition 5 nm thick (a common measure for a hard wall). The 5nm of protein is estimated to be 95% (± 5%) protein by volume from the measured refractive index of compressed layers. This approximates the volume of protein to be 2.96 * 10⁻⁸ cm³. A small protein such as Mefp-5 can be estimated to have a density of 1.45 g/cm^3 (Quillin et al.). This gives a film of 4.29 * 10⁻⁸ g of protein. A deposition of 20 µl of 20 µg/ml has 4 * 10⁻⁷ g of protein which gives a final deposition onto mica of ~11% the possible protein from bulk under these specified conditions.

In the adhesion measurements where the hard wall was 1 nm we can reasonably assume that we have 1/5 of the protein present at 5 nm. This gives us a deposition of 8.58 * $10^{-9} \frac{g}{\frac{1}{4}cm^2mica}$ and with a M_w of 9.5 kDa and assuming of the 20 mol of Dopa in Mefp-5, half (10) go to each of the two mica sheets, we average $3.61 * 10^{-7} \frac{mol \ Dopa}{m^2}$. If we take our strongest adhesion value of $0.0137 \frac{J}{m^2}$ we find that Dopa can be assigned $40.67 \frac{kJ}{mol}$. As each Dopa has 2 H-bonds to the mica substrate, a value of $20.34 \frac{kJ}{mol}$ seems quite reasonable and backs our assumptions as reasonable.

Supplemental Bibliography

Nozaki, Y., and Tanford, C. (1971) The solubility of amino acids and two glycine peptides in aqueous ethanol and dioxane solutions, *J. Bio. Chem.* 246, 2211–2217.

Quillin, M.L., and Matthews, B.W. (2000). Accurate calculation of the density of proteins. *Acta Crystallog. D Crystallography D 56*, 791-794.